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Microgravity Science and Applications Program Tasks

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I. INTRODUCTION

The all-encompassing objective of the NASA Microgravity Science and Applications (MSA) Program is the use of space as a laboratory to conduct research and development. The on-orbit microgravity environment, with its substantially reduced buoyancy forces, hydrostatic pressures, and sedimentation, enable scientists to conduct scientific investigations not possible on Earth. This environment allows processes to be isolated and controlled - and measurements to be made - with an accuracy that cannot be obtained in the terrestrial environment. Many of these processes also play dominant roles in diverse Earth-based technologies as well as technologies that support NASA's overall goals for future extraterrestrial exploration. The low gravity environment therefore demands investigations both for scientific and technological reasons. Examples of previous process research include growth of crystals and directional solidification of metals in the quiescent conditions in which gravitational fluid flow is eliminated; containerless processing of reactive materials to eliminate reactions with the container and to provide geometrical control of the product; and synthesis and separation of biological materials in weightlessness to reduce heat and mass transfer problems associated with sedimentation and buoyancy effects.

Emphasis will be placed on the expansion of currently funded activities for ground-based and space flight investigations to maximize the outputs from these opportunities. Initiatives requiring new hardware will be encouraged at a low level until funds can be made available. The expansion of current efforts is occurring as a result of focusing support for current space flight investigations on forming facility experiment teams to provide advice and identify future involvement. Emphasis has been placed on experiments to be flown in a series of Spacelab flights aboard the International Microgravity Laboratory (IML), the United States Microgravity Laboratory (USML), and United States Microgravity Payload (USMP).

The Microgravity Science and Applications Program Task Document covers the period of Fiscal Year 1991 (October 1990 - September 1991). The document includes research projects now being funded by the Office of Space Sciences and Applications, Microgravity Science and Applications Division, NASA Headquarters. The document is published annually and is sent to scientists in the field, both foreign and domestic. The information contained in this document is utilized in reports to the Associate Administrator, the Office of Management and Budget, and the Congress.

The Microgravity Science and Applications Division wishes to thank the Universities Space Research Association (USRA) and in particular Ms. Elizabeth Pentecost, for her efforts in the compilation and publication of this report.

II. TASKS

A. GROUND BASED EXPERIMENTS

1. ELECTRONIC MATERIALS

Process Modelling for Materials Preparation Experiments

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Center for Microgravity and Materials Research
Dr. J. Iwan D. Alexander
Dr. Jalil Ouazzani
Dr. Franz Rosenberger
NAG8-790 (NASA Contact: Dr. S. L. Lehoczky, MSFC)
August 1989 - July 1992

Objectives: The objectives of this program are the exploitation and improvement of available numerical techniques in order to formulate and solve useful models of transport processes in (microgravity) materials preparation experiments. Recognizing the fact that the potential of the most sophisticated modelling techniques cannot be fully realized if the physical properties, boundary and operating conditions are not well characterized, we aim to develop a comprehensive research program which coordinates a study of these essential ingredients with the development of numerical models for two specific crystal growth systems: (1) growth of mercury cadmium telluride (MCT) from the melt by the Bridgman-Stockbarger technique and (2) growth of triglycine sulfate (TGS) from solution.

Research Task Description: Experiments associated with this research include:

- o Transport property studies under representative crystal growth conditions (including undercooling and supersaturation):

Kinematic viscosity of molten mercury cadmium telluride as a function of temperature and composition.

Solute diffusivity in triglycine sulfate - water solutions.

- o Numerical modelling studies (using experimentally determined transport coefficients and boundary conditions):

The development of an algorithm which employs spectral techniques to solve a moving boundary problem. Subsequent incorporation of this technique into a pseudo-spectral collocation method that we have previously applied to convective-diffusive transport in melts. This will be followed by the development of numerical methods for the specific moving boundary problems associated with the growth of MCT by the Bridgman-Stockbarger technique.

Progress to Date:

The vapor pressure of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ in the compositional range of interest ($0 < x < 0.2$) lies between 15 and 40 atm. This, together with the relatively high melting temperatures involved, poses great difficulties for melt viscosity measurements with conventional techniques. Hence, we have developed a novel technique that is well suited to high pressure, high temperature conditions. The kinematic viscosity is obtained through gravimetric monitoring of the mass redistribution by melt flow, in a capillary that connects two sealed quartz chambers. This technique is highly accurate, as revealed by tests with room temperature liquids and low melting point metals of known viscosity. For melts with high surface tension, however, it is difficult to drive flow through small capillaries with small hydraulic heads. Hence, we have begun to develop in parallel a oscillating cup viscometer utilizing strain gauges.

Recent work has shown that the diffusivities of saturated and supersaturated solutions can be drastically lower than in the corresponding undersaturated solutions typically used for diffusivity determinations. The measurement of diffusivities of supersaturated solutions in diaphragm cells, that tend to stimulate nucleation, is difficult. Hence, we are developing a technique that minimizes nucleation stimulation. Diffusivities are obtained through the interferometric monitoring of the evolution of the concentration profile between two solutions of slightly different concentration. Though similar to the widely used Gouy technique, our approach does not require an initially sharp boundary between the solutions, and allows for a simpler evaluation of the interferometric data. Results obtained with highly concentrated NaCl solutions compare favorably with published values.

As a first step in the simulation of the solution growth of TGS, we have developed a 2-D model, with concentration-independent diffusivity and idealized heat transfer conditions, for the system used on Spacelab 3. The results show that, for the conditions of this experiment, diffusion dominated transport can prevail if the residual acceleration remains at or below a steady $10^{-6}g$ level. The suppression of convection at somewhat higher acceleration levels would require other operating conditions, such as lower cooling rates. The sensitivity to periodic acceleration perturbations was found to be complex.

Progress was also made toward the development of numerical codes for the MCT directional solidification problem. Employing a pseudo-spectral Chebyshev collocation technique, 3-D convection (without solidification) in a cylinder was treated. Steady and time-dependent moving boundary problems, involving heat conduction alone, have been solved, based on a 2-D Cartesian code developed earlier in our group. This approach has since been drastically modified to handle 2D axisymmetric situations. The method now hinges on a preconditioned conjugate gradient scheme which uses Chebyshev polynomials for the spatial discretization but employs a finite difference preconditioned operator to facilitate the solution of the problem. To date we have solved the moving boundary problem for heat diffusion only. We are currently examining the effects of finite ampoule wall thickness and will eventually incorporate the diffusing component. An axisymmetric (stream-function vorticity) code is being developed in parallel and will be used with the moving boundary code to solve solidification problems with melt convection.

Publications

Jin, W.-Q., Alexander, J. I. D. and Rosenberger, F., "Kinematic Viscosity Measurements at High Temperature and High Pressure - A Novel Technique," in preparation.

Nadarajah, A., Rosenberger, F. and Alexander, J. I. D., "Modelling the Solution Growth of TGS Crystals in Low Gravity," J. Crystal Growth **104**, 218 (1990).

Rosenberger, F., Alexander, J. I. D., Nadarajah, A. and Ouazzani, J., "Influence of Residual Gravity on Crystal Growth Processes," Microg. Sci. Tech. **III**, 162 (1990).

Pulicani J. P. and Ouazzani, J., " A Fourier-Chebyshev Pseudospectral Method for Solving Steady 3-D Navier-Stokes and Heat Equations in Cylindrical Cavities," Computers and Fluids, 1991 (n press).

Pulicani, J. P., Krukowski, S., Alexander, J. I. D., Ouazzani, J. and Rosenberger, F., "Convection in an Asymmetrically Heated Cylinder," International Journal of Heat and Mass Transfer, 1991 (accepted).

Alexander, J. I. D., Amiroudine, S., Ouazzani, J. and Rosenberger, F., "Analysis of the Low-Gravity Tolerance of Bridgman-Stockbarger Crystal Growth. II. Transient and Periodic Accelerations," J. Crystal Growth **113**, 21 (1991).

Rosenberger, F., "Boundary Layers in Crystal Growth, Facts and Fancy," in *Lectures on Crystal Growth* (H. Komatsu, ed.), 1991 (in press).

Growth of Zinc Selenide Crystals by Physical Vapor Transport in Microgravity

University of Alabama in Huntsville
Department of Physics
Dr. Elmer E. Anderson
Dr. Hai-Yuin Cheng
Dr. Franz Rosenberger
NAG8-767 (NASA Contact: Dr. S. L. Lehoczky, MSFC)
April 1989 - March 1992

Objectives: The overall objective of this program is to contribute to the understanding of the physical vapor transport (PVT) crystal growth method for possible future processing of II-VI compounds under low gravity conditions. As a basis, we strive to determine the optimum conditions for the PVT growth of single crystals of zinc selenide of good optical quality under normal gravity. A long-term objective is to extend the PVT method to the growth of mixed systems, such as ZnSe-ZnS. We aim to develop a research program with supplementary efforts in crystal growth, structural and optical characterization of the crystals, compositional characterization of the vapors and solids involved in the process, and in the numerical modelling of transport rates.

Research Task Description: Towards these objectives we are pursuing the following tasks:

- Vacuum conditioning and prepurification of starting materials to maximize transport rates. Since optimal transport rates can only be expected for quasi-congruent vapor composition, the starting material is preconditioned by a free sublimation that reduces volatile impurity concentrations and shifts in stoichiometry with respect to congruency.
- Vapor transport and crystal growth experiments in closed ampoules. This entails a study of nucleation distribution, transport rates and crystal habit as a function of ampoule dimensions and shape, and external temperature distribution.
- Vapor transport and crystal growth experiments in effusive ampoules, in order to provide for some uncoupling between transport rates and temperature distribution, and for further removal of incongruent vapor components during transport.
- Characterization of crystals by optical microscopy and scanning electron microscopy of etched and unetched crystal faces to determine the concentrations of dislocations, twin boundaries and possible precipitates. Infrared absorption spectra will be obtained for the best samples.
- Mass spectroscopic studies of the outgassing behavior of starting materials and silica ampoule material in order to provide guidance for the development of bake-out and seal-off procedures that will minimize the amount of impurities introduced into the vapor and, thus, potentially into the crystal.
- Numerical modelling of physical vapor transport for various conditions in temperature distribution, impurity or inert gas pressure, and vapor pressure differences between source and crystal interfaces. Our earlier PVT modelling and laser Doppler anemometry revealed geometrically rather complex transport flux distributions. Hence, realistic transport results can only be expected from 3-D models.

Progress to Date: The experimental setups for high-vacuum preconditioning, ampoule sealing, PVT crystal growth (including two three-zone furnaces, both with heat pipes), effusive ampoule vapor transport, etching and optical microscopy, mass spectroscopy (employing a high temperature effusion technique) have been completed and modified for the use with or the characterization of ZnSe.

About 26 growth runs in closed ampoules have been performed. With the evolution of optimal bakeout and preconditioning procedures for the source material, our transport rates in closed ampoules have recently increased to about 900 mg/day (18g/20 days). The crystal boule dimensions have increased to 2cm x 2.5cm, with only a few single crystalline grains per boule. While earlier boules showed extensive twinning and dislocation densities ranging from 10^4 - $10^7/\text{cm}^2$, a considerable reduction of defect densities can probably be achieved through more advantageous post-growth cooling programs. On samples where absorption spectra have been made, the IR transmission is greater than 80% from 1.8 to 15 microns. Electrical resistivity measurements indicated semi-insulating material.

Unseeded growth runs with the effusive ampoule technique were very promising and yielded growth rates of several 0.1 mm/hr. Currently, we are working on seeding procedures and an advantageous thermal geometry for large area seed plates.

The feasibility of the mass spectroscopic characterization of the outgassing products of silica ampoule material has been demonstrated. Initially, difficulties arose from an incompatibility between the BN effusion cell and the heating elements of the molecular beam epitaxy furnace used for sample vaporization. After containment of the SiO_2 samples in a platinum cell, that is insulated from the heating elements with electronic grade BN, runs of several days duration near the softening temperature of SiO_2 showed no deterioration of the effusion setup. Initial scans revealed drastic differences in the outgassing of different silica materials. In particular, flame fused silica glasses released at elevated temperatures large quantities of water vapor over days. This was associated with strong bubble formation in the bulk of the glass. Currently, we are concerned with a semi-quantitative characterization of the outgassing behavior, and the development of high-vacuum pre-annealing procedures that will reduce the outgassing during subsequent use of the container material for crystal growth.

In the 3-D modelling of PVT rates we have encountered great difficulties. A comparison of numerical results with transport rates obtained in our group under well characterized conditions, revealed the inability of the commercial code used to produce accurate results. After extensive interaction with the developer of another code, and the running of some benchmark cases, we believe, that we will be able to satisfactorily tackle this problem in the near future.

Memory Effects in Organometallic Molecular Beam Epitaxy

North Carolina State University
Professor Klaus J. Bachmann

In the second year of funding the work on chalcopyrite structure wide gap materials was continued adding research on group IV-wide gap III-V heterostructures. Two systems $\text{Ge}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ and Si/GaP were considered. The research on $\text{Ge}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ involved a collaboration with Dr. M. Timmons and Dr. J.B. Posthill of the Research Triangle Institute and focussed on the trade-off between improvements in the minority life time in the epitaxial $\text{Al}_x\text{Ga}_{1-x}\text{As}$ on the germanium substrate and the degradation of the interfacial recombination velocity with increasing substrate temperature which is optimum at $\sim 700^\circ\text{C}$. The work on Si/GaP focussed in accord with the program goal of replacing highly toxic and pyrophoric source materials for chemical beam epitaxy and metalorganic chemical vapor deposition by inert solid sources, on the identification of a solid source for phosphorus replacing the need for storage of phosphine in pressurized containers. We were successful of using solid red phosphorus that was heated to temperatures in the range $250\text{--}400^\circ\text{C}$ to inject phosphorus vapor into a remote hydrogen plasma from which then highly reactive phosphorus hydride species (PH) could be extracted into a vacuum chamber combining on a heated silicon or GaP substrate with downstream injected $\text{Ga}(\text{CH}_3)_3$ to form a GaP epitaxial film. This process works at substantially lower substrate temperature ($<650^\circ\text{C}$) than conventional oxygen chemical vapor deposition (MOCVD) of GaP which proceeds typically at $800\text{--}900^\circ\text{C}$. A remaining problem is the relatively high carbon concentration in the GaP that is doped p-type requiring further research to control this problem. The research on III-V/IV heterostructures is connected to the first year research on II-IV-V₂ alloy heterostructures by the discovery of a wide range of metastable solid solutions between the II-IV-V₂ and group IV elements. This opens the intriguing possibility of continuously grading from the diamond structure into the chalcopyrite structure maintaining exact lattice matching to silicon through the entire structure and terminating at a polar surface entirely occupied by phosphorus atoms. This surface could serve as template for the growth of other compound semi-conductors, such as $\text{Al}_x\text{Ga}_{1-x}\text{P}$ without the nucleation problems that plague the growth of such films directly on silicon. Since the pay-offs of such a technology would be substantial, we will pursue $\text{Si}/\text{Al}_x\text{Ga}_{1-x}\text{P}$ as a next step of this program. Also, we will continue with the development of solid sources for molecular beam epitaxy (MOMBE) and MOCVD.

Publications

Xing, G.C., Bachmann, K.J., Posthill, J.B., and Timmons, M.L., "Substrate Effects on the Epitaxial Growth of ZnGeP_2 Thin Films by Open Tube MOCVD," *J. Appl. Phys.* **69**, 4286 (1991).

Xing, G.C., Posthill, J.B., and K.J. Bachmann, "Organometallic Chemical Vapor Deposition and Characterization of $\text{ZnGe}_{1-x}\text{Si}_x\text{P-Ge}$ Alloys on GaP Substrates," *J. Cryst. Growth* **113**, 113 (1991).

Choi, S.W., Bachmann, K.J., Posthill, J.B., and Timmons, M.L., "The Growth and Characterization of $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{Ge}$ Heterostructures," *J. Electrochem. Soc.*, 1991 (accepted).

Bachmann, K.J., "Chalcopyrite Structure Semiconductors", in *A Concise Encyclopedia of Electronic Materials* (S. Mahajan, ed.), Pergamon Press, 1991 (in press).

Fluid Mechanics and Mass Transfer in Melt Crystal Growth: Macro- and Micro-Scale Analysis of Controlled Solidification

Massachusetts Institute of Technology
Professor Robert A. Brown
NAS 7645 (NASA Contact: Mark Lee)
October 1, 1989 - September 31, 1992

Objectives: This program is aimed at fundamental understanding of the interactions of heat and mass transfer, melt flow and interface morphology in the design and interpretation of solidification experiments for growth of crystals on earth and in space. The ongoing research is a mixture of state-of-the-art computational studies of macroscale transport in large-scale crystal growth systems and of microscale formation of cellular solidification structures. An experimental program for studying mechanisms for microstructure formation in solidification and is support by theoretical and computational analysis of cellular and dendritic crystal growth of binary alloys.

Research Task Description: The research being performed under this program is divided into several distinct tasks according to the application of the research to the understanding of either macroscale or microscale features in solidification. These are as follows:

Simulation of Directional Solidification. Application of transient and steady-state calculations of axisymmetric convection in directional solidification systems to analysis of both vertical gradient freeze and Bridgman-Stockbarger configurations for growth of semiconductor and oxide crystals. This project includes development of the simulators to include detailed models for radiative heat transfer and coupling between the temperature, solutal and convection fields.

Cellular and Dendritic Crystal Growth in Thin-film Solidification. Experimental and computational analysis of the dynamics of cellular interface formation in thin-film, two-dimensional directional solidification of a binary alloy. Development of a new numerical method for analysis of the transition from shallow to deep cellular surfaces is included.

Simulation of Small-Scale Floating Zones. Simulation of heat transfer, convection and interface morphologies in small-scale floating zones. Analysis should predict the onset of instabilities in the zone caused by both surface-tension-driven convection and by interactions of the zone shape with heat transfer.

Three-dimensional Convection in Solidification. The development of the first numerical method for analysis of flow and interface morphology with three-dimensional convection in directional solidification. This research involves the development of numerical algorithms for solving three-dimensional convection problems with solidification which are robust on medium-grain parallel processing computers.

Progress to Date: The progress to date in each of these projects is summarized here. Publications that describe these developments are listed below.

Simulation of Directional Solidification. The simulation for heat transfer, convection, dopant transport and interface morphology in directional solidification has been applied to the analysis of vertical Bridgman growth of HgCdTe and the gradient freeze system for growth of GaAs crystals. In both systems experimental observations of the melt/crystal interface morphology and of dopant segregation are reproduced and design issues that impact dopant uniformity are addressed. The simulator has also been used to explore the usefulness of the concept of the effective segregation coefficient for axial segregation in directional solidification. Bounds for the accuracy of this concept have been established.

The simulation for directional solidification has been extended to include a complete analysis of diffuse gray in the furnace; this extension makes possible design calculations that connect the furnace configuration with crystal quality through dopant segregation and defects in the crystal.

Cellular and Dendritic Crystal Growth in Thin-film Solidification. Progress in understanding cellular solidification has come in both theoretical and experimental parts of the program. First, calculations of the transition from shallow to deep solidification cells has demonstrated conclusively that there is no mechanism for selection of cells of a unique wavelength, based on the existence of steady-state solutions. Moreover, the first fully dynamical simulations of collections of cells have shown that multiple steady, time-periodic and apparently chaotic states are possible. Also, the apparent wavelength of the cells decreases very rapidly with increasing growth rate through a tip splitting mechanism that is connected with the codimension-two bifurcation between cell shapes with spatially resonant wavelengths. A large-scale thin-film solidification system has been developed for studying the dynamics of thin-film solidification of succinonitrile-acetone alloys. The system allows video imaging and image processing of a large collection of cells for statistical analysis of cellular dynamics. Results obtained in the system agree qualitatively with the theoretical predictions; chaotic dynamics are observed that involves a band of wavelengths which migrates to lower mean values as the growth rate is increased.

Simulation of Small-Scale Floating Zones. A complete simulation of axisymmetric convection, heat transfer and interface morphologies in small-scale floating zone systems has been developed for solidification of binary alloys. This simulation is being extended to the analysis of the stability of these steady-state flows to nonaxisymmetric disturbances to look for the onset of the traveling wave states caused by surface-tension-driven convection.

Three-dimensional Convection in Solidification. The finite-element/Newton method developed by this research group for solution of the coupled convection/solidification problems is being adapted for use on medium-grain parallel computers; our first calculations are on an Intel i860 hypercube with 32 vector computational elements. Initial development work has focussed on the development of direct development of domain decomposition, incomplete nested dissection method for LU decomposition of the large sparse asymmetric matrices that arise as the Jacobian matrices that arise in the Newton iterations using finite-element methods. The development of the algorithm has been completed and gives calculations that make use of over 80 percent of the computer power of a 16-processor Intel i860 hypercube and give computational times very similar to a single processor Cray Y-MP processor. The algorithm scales linearly with the number of processors and so is amenable to much faster execution times on larger MIMD parallel machines. We are testing this hypothesis.

Publications

Kim, D.-H. and Brown, R.A., "Transient simulation of convection and solute segregation of GaAs growth in a vertical gradient freeze furnace," *J. Cryst. Growth* **109**, 66-74 (1991).

Brown, R.A. and Kim, D.H., "Modelling of directional solidification: from Scheil to detailed numerical simulation," *J. Cryst. Growth* **109**, 66-74 (1991).

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Lee, C.T., Ramprasad, N. and Brown, R.A., "Two-dimensional cellular structures in the solidification of $\text{CBr}_4\text{-Br}_2$: comparison of calculations with experiments," *J. Cryst. Growth*, 1991 (submitted).

Bennett, M.J. and Brown, R.A., "Nonlinear Dynamics in Periodically Repeated Sets of Directional Solidification Cells," Phys. Rev. B., 1991, submitted.

Brown, R.A. "Perspectives on Integrated Modeling of Transport Processes in Semiconductor Crystal Growth," in *Advanced Heat Transfer in Manufacturing and Processing of New Materials*, Hemisphere Press, 1991 (in press).

Lee, J.T.C. and Brown, R.A. "The Planar to Cellular Transition and the Long Time Scale Dynamics of Pattern Formation During Thin-Film Directional Solidification," *Materials Research Society*, 1991 (in press).

Krylidis, A. and Brown, R.A., "On the Ability of Nonperturbative Density Functional Theories to Stabilize Arbitrary Solids," Phys. Rev. A, 1991 (in press).

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Tsiveriotis, K. and Brown, R.A., "Solution of Free-boundary Problems using Finite-element/Newton Methods and Locally Refined Grids: Application to Analysis of Solidification Microstructure," Int. J. Num. Meth. Fluids, 1991 (submitted).

Kyrlidis, A. and Brown, R.A., "Spatially Varying GELA (SVGELA) Formulation of Density Functional for Nonhomogeneous Hard Sphere Systems," Phys. Rev. B, 1991 (submitted).

Lee C.T. and Brown, R.A., "Experimental Investigation of Nonlinear Dynamics near the Planar to Cellular Transition in Thin-Film Directional Solidification," Phys. Rev. A, 1991 (submitted).

Modeling Internal Radiative Transport in Crystal Growth Processes

University of Minnesota
Professor Jeffrey J. Derby

Objectives: The goal of this research program is to understand the effects of internal radiation in several crystal growth systems of technological importance. We will develop new, robust methods to accurately predict radiation transfer in semitransparent media for the complicated, multi-dimensional geometries of crystal growth systems. The methods developed for describing radiation transport will be combined with previous heat transfer and hydrodynamic analyses of crystal growth to provide quantitative, predictive capabilities for these processes.

Radiation energy transfer is an important feature in the high-temperature processes used to produce many optical and semiconductor crystals. The impact of radiative energy exchange among *surfaces* is universally recognized in melt growth systems; however, the effect of internal radiation transport *within* participating solid and liquid phases is not well understood. Internal radiation heat transfer is significant during the melt growth of many crystals used in optical and photoelectronic devices.

Progress to Date: We have successfully developed a Galerkin finite element method for solution of the integro-differential equations which describe combined-mode heat transfer problems. This method is very accurate for complicated geometries and all values of optical thickness. We have applied these methods to a model of the vertical Bridgman growth of semitransparent crystals. Details of this work can be found in the listed references.

Perhaps the most interesting results are those which indicate the strong influence of internal radiation on the shape and position of the melt/crystal interface and the temperature gradients surrounding it. Heat transfer through the system is strongly affected by the optical absorption coefficient of the crystal. Coupling of internal radiation through the crystal with conduction through the ampoule walls promotes melt/crystal interface shapes which are highly deflected near the ampoule wall. This "radiative interface effect" is much more pronounced than that observed in the Bridgman growth of opaque crystals, where the interface deflection at the ampoule wall is attributed to the thermal conductivity mismatch between ampoule and charge. Calculations demonstrate that a flatter overall interface shape can be achieved through optimization of ampoule material properties and furnace temperature profiles.

The primary disadvantage of this algorithm is the significant computational expense needed for accurate calculations. This feature has limited our attention to the vertical Bridgman system, where the relatively simple geometry is used in the calculations. We are exploring the use of parallelism to speed our algorithm. We believe that this methodology will enable further studies of the vertical Bridgman system and analysis of the more complicated Czochralski crystal growth system.

Publications

Brandon, S. and J.J. Derby, "Internal Radiative Transport in the Vertical Bridgman Growth of Semitransparent Crystals," J. Cryst. Growth **110**, 481--500 (1991).

Brandon, S. and J.J. Derby, "A Finite Element Method for Internal Radiative Heat Transfer and its Application to Analysis of the Growth of Semitransparent Crystals," in *Fundamentals of Radiation Heat Transfer*, HTD-Vol. 160 (W.A. Fiveland, A.L. Crosbie, A.M. Smith, and T.F. Smith, EDS.), ASME, 1991, pp. 1--16.

Brandon, S. and J.J. Derby, "Heat Transfer in Vertical Bridgman Growth of Oxides: Effects of Conduction, Convection, and Internal Radiation," *J. Cryst. Growth*, submitted (1991).

Brandon, S. and J.J. Derby, "Finite Element Methods for Analysis of Internal Radiative Heat Transfer and Solidification in a Finite Cylindrical Enclosure," *Int. J. Numer. Meth. Engng.*, 1991 (submitted).

Presentations

Brandon, S. and Derby, J.J., "Internal Radiative Heat Transfer during the Growth of Oxide Single Crystals," *Eighth American Conference on Crystal Growth, ACCG-8*, Vail, CO, July 15-20, 1990.

Brandon, S. and Derby, J.J., "Internal Radiative Heat Transfer during the Growth of Oxide Single Crystals," *American Institute of Chemical Engineering Annual Meeting*, Chicago, IL, November 12-16, 1990.

Brandon, S. and Derby, J.J., "Internal Radiative Heat Transfer during the Growth of Oxide Single Crystals," *Twelfth Conference on Crystal Growth, AACG/west-12*, Fallen Leaf Lake, CA, May 28-31, 1991.

Derby, J.J., Salinger, A.G., Brandon, S., and Aris, R., "Multiple Flows of a Radiatively Participating Fluid in a Finite Cylindrical Enclosure: A Finite Element Analysis of a Rayleigh-Benard Problem with Internal Radiation," to be presented at *American Institute for Chemical Engineering Annual Meeting*, Los Angeles, CA, November 17-21, 1991.

Derby, J.J., "Macroscopic Interactions of Viscous Flow, Heat Transfer, and Solidification Interfaces in Melt Crystal Growth Systems: Oxides and Fluorides," *IMA Workshop: Free Boundaries in Viscous Flows*, Minneapolis, MN, March 11-15, 1991.

Brandon, S. and Derby, J.J., "On the Analysis of Internal Radiative Transport during the Growth of Semitransparent Crystals," poster presentation at *Gordon Research Conference for Crystal Growth*, Plymouth State College, Plymouth, NH, July 15-19, 1991.

Derby, J.J., "Computer-Aided Analyses of Internal Radiation and Convection: Applications to Crystal Growth and Glass Processing," *Laser Division Seminar Series*, Lawrence Livermore National Laboratory, Livermore, CA, September 17, 1991.

Derby, J.J., "Computer-Aided Analyses of Internal Radiation, Conduction, and Convection in Crystal Growth Processes," *James and Catherine Patten Seminar series*, Department of Chemical Engineering, University of Colorado at Boulder, Boulder, CO, September 19, 1991.

Crystal Growth by Two Modified Floating-Zone Processes

University of Wisconsin

Professor Sindo Kou

NAG8-705 (NASA Contact: S.L. Lehoczky, MSFC)

March 1, 1988 - February 28, 1991

Objectives: The objectives of this study are: (1) to help reduce melt-zone convection in floating-zone crystal growth under microgravity; and (2) to help control crystal cross-sectional geometry. Floating zone crystal growth under microgravity, which although essentially free from natural convection, can still suffer from thermocapillary (Marangoni) convection. It has been reported that oscillatory thermocapillary convection can cause dopant striations in the resultant crystals.

Research Task Description: To effectively reduce this convection while at the same time help produce single crystals of a uniform diameter and smooth surface, two modified floating-zone processes have been developed. In these processes a shaper is used to define the cross-sectional shape of the growing crystal and to reduce the area of the free surface. As a result, crystals with a very uniform diameter and smooth surface can be grown, and thermocapillary convection can be dramatically reduced. In modified process 1 the shaper is in the form of a ring covering most of the free surface, whereas in modified process 2 it is in the form of a step immersed in the melt. The temperature of the shaper is carefully controlled during crystal growth. The approach is as follows: (1) Flow visualization using a transparent melt in which Marangoni (rather than natural) convection dominates so as to allow melt-zone convection under microgravity to be simulated in ground-based experiments; (2) Crystal growth and examination of cross-sectional variation along the crystal axis; and (3) Computer simulation using general body-fitting curvilinear coordinates, allowing consideration for shapes of unknown melt/crystal interfaces.

Progress to Date: (1) Flow visualization using the NaNO_3 melt has demonstrated much less convection in both modified processes; (2) Computer simulation confirms flow-visualization experiments; (3) Growth of NaNO_3 crystals shows good cross-sectional control; and (4) Computer simulation of both modified processes has demonstrated the dramatic reduction in thermocapillary convection.

Publications

Kim, Y.J. and Kou, S., "Thermocapillary Convection in Zone Melting Crystal Growth - An Open Boat Physical Simulation," J. Cryst. Growth **98**, 637 (1989).

Lan, C.W., Kim, X.J., and Kou, S., "A Half-Zone Study of Marangoni Convection in Floating-zone Crystal Growth under Microgravity," J. Cryst. Growth **104**, 801 (1990).

Lan, C.W. and Kou, S., "Thermocapillary Flow and Melt/Solid Interfaces in Floating-zone Crystal Growth under Microgravity," J. Cryst. Growth **102**, 1043 (1990).

Lan, C.W. and Kou, S., "Floating-zone Crystal Growth with a Heated and Immersed Shaper - Computer Simulation," J. Cryst. Growth **108**, 340 (1991).

Lan, C.W. and Kou, S., "Floating-zone Crystal Growth with a Heated Ring Covering the Melt Surface," J. Cryst. Growth **108**, 1 (1991).

Lan, C.W. and Kou, S., "Floating-zone Crystal Growth with a Heated and Immersed Shaper - Experiments," J. Cryst. Growth **108**, 541 (1991).

Lan, C.W. and Kou, S., "Thermocapillary Flow and Natural Convection in a Melt Column with an Unknown Melt/Solid Interface," Int. J. Num. Meth. Fluids **12**, 59 (1991).

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Theory of Materials Growth in Space-The Liquid-Solid Interface

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NAS1-18226

Objectives: The objective of this research is to advance the theory to improve our knowledge of the detailed phenomena occurring near the liquid-solid growth interface.

Research Task Description: In this project, we plan an accurate evaluation of many-body interactions on growth surfaces in the presence of nonidealities such as reconstruction, native defects, alloying, and non-isoelectronic defects. Using the calculated energies, the equilibrium free energy will be found for metal-liquid interface. Growth rates will be studied by solving appropriate kinetic equations.

Progress to Date: Many body interactions in the bulk and on several surfaces have been obtained with a fitted tight-binding (TB) Hamiltonian in the generalized perturbation method (GPM). As a test case, the calculations were carried out for GaAs. This method has also been successfully used to study surface-driven ordering in GaAlAs alloys.

The empirical bandstructures, even when they are carefully constructed for the cases studied, are limited in versatility and accuracy, while most of the existing first principle bandstructures that employ LAPW, or LMTO, or pseudopotential method with plane waves are computationally too demanding to study realistic problems. One way to overcome this hurdle is to use the linear combination of atomic orbitals (LCAO) method in a TB form. This first-principles tight-binding Hamiltonian (FPTB) uses pseudo-atomic-orbitals as the basis for a self-consistently determined Hamiltonian matrix. These orbitals are computed self-consistently from a free-atom calculation within LDA. A major advantage of LCAO method over other methods is its availability in the familiar TB form which is perfectly suitable for all the tasks to be studied under this project. Particularly, incorporation of this Hamiltonian in the Green's function approach either to study surface-driven ordering, or to obtain coherent potential approximation limited alloy band structure which is used to calculate many-body energies in a GPM, or to study the effect of impurities and defects on bandstructures is straightforward. We are currently involved in the development of this method.

We have developed the programs for the calculation of the ternary phase diagrams within the quasichemical approximation. Good agreement for well studied systems, such as InGaAs, is found. Calculations to date are based on pair wise interactions and mixing energies. We are currently extending the calculations to include energies for larger clusters, more specifically a 16-bond cluster. These calculations should improve the agreement with experiment, in particular in cases where the pairwise approximation is a poor one.

Growth Kinetics of Physical Vapor Transport Processes. Crystal Growth of an Opto-Electric Material: Mercurous Chloride

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NAS3-25274 (NASA Contact: Dr. Walter Duval, LeRC)

Objectives: Halides of mercury (I) are extremely interesting compounds because of their large birefringence, wide range of transparency and high acousto-opto figure of merit. No commercially available material exhibits the broad transmission range, high figure of merit and slow sound velocity of these halides. The ultimate goal is to produce very high quality Hg_2Cl_2 crystals which have many advantages in device over existing commercial material such as quartz, TeO_2 , PbMoO_4 , and LiNbO_3 .

Mercurous chloride crystals are grown by Physical Vapor Transport (PVT) method in a sealed quartz tube. To be practical as an optical signal processing device, an acoustic cell must be made from material which does not scatter the optical beam as it passes through the crystal. Our experiments on continuous improvement in scattering reduction showed that low scattering and increased optical homogeneity can be achieved under reduced thermal and solutal convection conditions. The overall objective of this program is to clearly identify the growth condition under which crystals with uniform refractive index could be grown. The control of convection during the crystal growth will maximize the chemical and optical homogeneity which ultimately provide low optical and acoustic attenuation.

Research Task Description: Experiments were carried out to derive a quantitative understanding of the physical vapor transport process, in order to identify the magnitude of convection effects on the growth process. Effects of convection on the crystal quality were studied by varying the growth system aspect ratio and thermal conditions which ultimately influence thermal convection during the vapor transport process. In addition, kinetic measurements were made for the PVT process as a function of g-vector studies. The aspect ratio and thermal parameters were kept identical for all the g-vector. The change in convection conduction was only due to variations in g-vector.

Progress to Date: The crystal growth rate increased with increasing convection level up to a certain value and then dropped to a constant level, indicates a very complex transport geometry which cannot be explained by the linear stability theory. The better quality of crystal grown at a low Rayleigh number confirmed that improved optical properties are possible in convectionless environment. Results of growth velocity measurements obtained at different g-vector (at the studied $0t$) showed significant variations in the growth velocity. This suggests that strong convective flows are present. This work is still in progress. The preliminary results are supported by findings of Mennetrier, Duval and Singh where effect of g-vector has been modeled for this configuration for gravity levels from 10^{-6} g to 1-g. Purely diffusive flow and no recirculating cells were predicted below 10^{-3} g for mercurous chloride crystal growth indicating the value of microgravity environment of crystal growth.

Publications

Singh, N.B., Gottlieb, M., Brandt, G.B., Nicholson, D., Davis, J., Blanchard, D., Glicksman, M.E., Singh, M., and Paradies, C., "Stoichiometric Considerations during Crystal Growth of Mercurous Halides," Mat. Lett. **11**, 31 (1991).

Singh, N.B., Hopkins, R.H., Mazelsky, R., and Glicksman, M.E., "Effect of Convection on the Physical Vapor Transport Growth of Opto-electronic Crystals," in *Proceedings of CSME* (I.G. Currie, ed.), 1991 (to be published).

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Singh, N.B. and Duval, W.M.B., "Flight Experiment on Acousto-optic Crystal Growth," in *Proceedings of AIAA/IKI Microgravity Science Symposium*, AIAA, 1991, p. 179.

Presentations

Singh, N.B., "Recent Advances in Photonic Crystals," *Mid-Atlantic Section of AACG*, 1991.

Singh, N.B., "Flight Experiments on Crystal Growth," *AIAA Committee Meeting*, Pasadena, 1991.

Vapor Crystal Growth of Electrooptical Materials

NASA Marshall Space Flight Center

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Dr. S. L. Lehoczky

Dr. C.-H. Su, USRA

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In-House and with Boeing

Continuing task

Objectives: The goal of this project is to establish the effects of the processing environment, including gravitational orientation and nature and distribution of defects on materials of substantial technological interest that can be grown by the physical vapor transport method. The primary candidate materials of this project are CdS and CdTe.

Research Task Description: The investigation is intended to define a flight experiment that will take full advantage of the vapor crystal growth furnace being developed by Boeing Aerospace Company. That furnace has been made available to Marshall Space Flight Center (MSFC) for one flight. The strategy includes the development of a 1-g crystal growth data base and extensive characterization of both the starting materials and the grown crystals. A method to simulate reduced gravity has not been devised for this growth method but crystals will be grown at several orientations with respect to the gravity vector to assess the effects of gravity to the greatest extent possible on Earth. The material for the first flight, currently planned for mid-1992, will be CdTe.

Progress to Date: Favorable growth parameters in a laboratory furnace have been established. Work with Boeing was begun in the Boeing laboratory and ground control furnaces in Seattle. Additional growth runs will be necessary to define the optimum flight parameters. The University of Alabama in Huntsville has analyzed both ampoule and sample materials and results indicate that adequate purity for achieving the project objectives exists in the starting materials and is maintained during the CdTe compounding process. Purity of CdTe crystals following the growth process will be determined. In addition to CdTe and CdS, high quality crystals of ZnTe, PbS, and PbSe have been grown in the laboratory at MSFC using the technique developed under this project. The ZnTe crystals grown under this project have optical transmission and photoluminescence spectra that are featureless, as opposed to various bands observed in the spectra of ZnTe grown by the travelling heater method (THM).

Publications

Su, C.-H., Lehoczky, S. L., and Szofran, F. R., "An Improved Vapor Growth Method for the Preparation of Electronic and Electro-Optical Materials," NASA Tech Brief, 1991 (in press).

Volz, M. P., Su, C.-H., Lehoczky, S. L., and Szofran, F. R., "Vibronic Spectra of Cu²⁺ in ZnTe," Phys. Rev. B, 1991 (submitted).

Solution Crystal Growth of Organic and Polymetric Materials for Nonlinear Optics Applications

NASA Marshall Space Flight Center

Dr. Marcus Vlasse,

Dr. Donald O. Frazier, MSFC

Dr. H.J. Caulfield, University of Alabama, Huntsville

Dr. S. C. Narang, Stanford Research Institute

Objectives: The major objective of the research is the thorough ground-based investigation of the basic aspects of solution growth of several representative diacetylenes and other organic compounds with nonlinear optical properties. Among the aspects to be studied are the influence of solvents, thermal gradients, concentration gradients, and, foremost, the influence of convective flows on the growth process and perfection of the crystal. Further problems to be studied are the evaluation of the influence of interface effects on defect incorporation into the crystals. Size and shape and growth habit of the crystal may play important roles in the growth process.

The results of the study will be used to determine the advantages of growing such crystals in space under a reduced gravity environment. Furthermore, these findings will be useful in solving current problems in the crystal growth of organic or biorganic substances with nonlinear optical properties.

Research Task Description: In this ground-based crystal growth experiment, the plan is the control of convection in a predictable manner as to be able to measure its effects and extrapolate to microgravity environment.

The following basic steps will form the bulk of the research task:

A small scale growth experiment will be carried out using the evaporation method to produce crystals for further study and seeding. The bulk of the work will be performed using the controlled cooling method to produce the necessary supersaturation and induce nucleation and growth. Both seeded and unseeded runs will be introduced into this growth method to determine its effect on the growth habit. Reduction of convection will also be tried by the use of more viscous media such as gels, where the process is quasidiffusion controlled. The geometry of the growth cell and the position of the seed in this cell will be used as a major means in reducing convection effects.

Detailed characterization of the crystals will be performed to determine the densities of chemical and physical impurities. Such characterizations will be performed mainly by spectroscopic means as well as microscopy and x-ray diffraction and topography. These data will be correlated to the various growth parameters and methods in order to determine the influence of such parameters on the quality and size of crystals. Some simple modeling of the growth process will be attempted to acquire a better understanding of the kinetics and mechanisms involved.

Progress to Date: The apparatus and method have been set up for the preparation of a urethane substituted diacetylene ($R-C\equiv C-C\equiv C-R$), TCDU, not available in commerce.

The conditions (solvents, temperature, saturation, etc.) have been investigated for the growth of unseeded and seeded crystals of TCDU by the evaporation method. Single crystals of TCDU, in the form of plates of good optical quality have been grown from ethyl acetate-ethanol mixtures at room temperatures.

Work has been initiated to study the crystal growth of L-arginine phosphate (LAP), a very promising material for NLO applications. The dependence of its solubility on temperature and pH in aqueous solutions has been determined. Saturation and speed of evaporation have been studied. Seeded

growth experiments in various configurations have been performed. The results of this work so far have allowed us to grow single crystals of LAP of extremely good optical quality for further experimentation.

The apparatus for the application of controlled and programmable cooling has been set up and the cooling regimes have been determined. Temperature stability of 0.1°C has been attained. Growth trials using this method have started. The results of several trials on LAP crystals using qualitative microscopic observations of surface roughness and growth step size show that the geometric position of the seed and the rate of saturation are two of the main contributors to crystal quality. These factors will now be quantified. This growth method, apparatus and growth parameters will lead eventually to a possible flight experiment.

Using the above apparatus experiments were carried out using a stable geometric configuration of the growth environment (isothermal downward crystal growth) to offset the effects of buoyant convection and produce a minimal convection regime. This approach has produced crystals of differing quality depending on the growth direction. The downward grown crystals are large, clear, smooth, with no optical defects and no secondary nucleation. The upward grown crystals are small, rough surfaces with many cracks and defects and accompanied by secondary growth.

Publications

Vlasse, M. and Mitchell, T., "Solution Crystal Growth in a Simulated Space Environment," J. Cryst. Growth, 1991 (submitted).

Modelling Directional Solidification

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M. Banan, Co-I
J. Zhou, Co-I
R. Gray, Co-I
W. Yuan, Co-I
NAG8-831 (NASA Contact: F. Szofran, MSFC)
May 1, 1990 - April 30, 1993

Objective: The objective is to gain an improved understanding of the influence of gravity on the directional solidification of compound semiconductors and their alloys.

Research Task Description: The following tasks have been undertaken under the current grant.

1. Determine experimentally the influence of current pulses and vibration on compositional homogeneity and microstructure of InSb-GaSb alloy.
2. Determine experimentally the variation of freezing rate during a spin-up/spin-down cycle by using current pulses.
3. Determine experimentally the influence of vibration and ACRT on the interface shape of doped InSb using current pulses during solidification.

Progress to Date: Task 1 has been completed. Tasks 2 and 3 are underway.

The variation of In/Ga with distance down the ingot corresponded to complete mixing in the melt, regardless of whether current pulses, vibration, or spin-up/spin-down was employed or not. This indicates that there was sufficient buoyancy-driven convection in this apparatus to completely mix the melt at the low freezing rates required to avoid constitutional supercooling. The number of grain boundaries and twins was increased by application of current pulses, but decreased by vibration of the ampoule. There seemed to be an optimum rotation rate for minimal grain size with ACRT. We suspect that the primary effect of vibration and ACRT is to make the interface shape more convex, thereby aiding grain selection.

Doped InSb is being used with current interface demarcation to determine the microscopic variation of freezing rate. Striations were produced both by the current pulses and by starting and stopping the ampoule rotation. The freezing rate varied during the ACRT cycle. Thus far we have seen no evidence of melt back.

A technique was developed for measuring the heat transfer coefficient between the material in an ampoule and the furnace wall. This coefficient has been used in many theoretical treatments of heat transfer in the directional solidification, but until now there was no means for measuring it.

Publications

Banan, M., Gray, R.T., and Wilcox, W.R., "A Experimental Approach to Determine the Heat Transfer Coefficient in Directional Solidification Furnaces," J. Cryst. Growth 1991 (submitted).

Presentations

Zhou, J., Banan, M., Gray, R., and Wilcox, W.R., "Use of Current Interface Demarcation during Directional Solidification of Te-doped InSb with ACRT," *Third Canadian Materials Science Conference*, Toronto, June 1991.

Wilcox, W.R., "Gravitational Effects on Directional Solidification of Indium Antimonide and Indium Gallium Antimonide," University of Colorado, Boulder, August 1991.

Zhou, J., Banan, M., Gray, R., and Wilcox, W.R., "ACRT Striations and the Use of Current Interface Demarcation during Directional Solidification of Te-doped InSb," *ACCG/East-4*, Atlantic City, October 1991.

Crystal Growth of Device Quality GaAs in Space

Massachusetts Institute of Technology
Professor August F. Witt
NAGW-1563

Research during the past funding period was directed at:

(a) Growth Control in Bridgman and Gradient Freeze Geometries

In line with advanced rate control concepts developed during past funding period, a vertical Bridgman system with heat-pipe based heat transfer control was redesigned, reconstructed and put into operation. Structural modifications were directed at establishing compatibility of our hot zone with the Crystal Growth Furnace (CGF) facility. Conventional PID controllers were eliminated and control functions transferred to the Masscomp computer facility. A drive mechanism for axial displacement of the control sensor was installed; current induced growth interface demarcation capability was established. Growth experiments with the pseudo-binary system GaInSb are performed to establish quantitatively the prevailing thermal boundary conditions.

(b) Design of Bridgman Geometry for Growth of Bismuth Silicate and Germanate

In conjunction with the design of a hot zone for Bridgman-type growth of nonlinear optical oxides, numerical analysis of the effects of crucible materials and structure on growth interface morphology was conducted. It was found that conductance along the crucible wall and the degree of thermal coupling between furnace and crucible are dominant parameters. In this respect oxide growth in Bridgman configuration differs fundamentally from that of conventional semiconductors and requires alternate hardware design and control features. These are currently being pursued.

(c) New Optical Approaches to the Quantitative Characterization of Growth and Defect Structures in Semiconductors

Using NIR transmission microscopy with computational absorption analysis, the effects of axial magnetic fields on macro- and micro-segregation during LEC growth of GaAs were quantitatively investigated with a spatial resolution of close to 2 μm . Segregation inhomogeneities in excess of one of magnitude are found to be related to fluid dynamics of the melt. The applicability of the BPS theory as well as the non-applicability of the Cochran analysis were established.

Publications

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2. SOLIDIFICATION OF METALS, ALLOYS, AND COMPOSITES

Immiscible Phase Incorporation During Directional Solidification of Hypermonotectics

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Dr. R. T. Dehoff, University of Florida, Co-I

NAG8-768 (NASA Contact: Dr. P.A. Curreri, MSFC)

Objectives: The objective of this work is to investigate morphological developments during the directional solidification of immiscible (hypermonotectic) materials. One of the most important factors in developing immiscible alloys for eventual applications involves ways to control the microstructure of the alloys through solidification. For most applications, it is desirable to have a larger amount of the immiscible phase in the final structure than that provided by an alloy of monotectic composition. A higher volume fraction can be reached by using a higher solute content (i.e. hypermonotectic) alloy. However, sedimentation and convective instabilities can result in difficulties during processing. This research is attempting to answer questions concerning the events which occur at the solidification front during directional solidification of hypermonotectic alloys. In particular, it is desirable to determine the mechanisms behind the events observed.

Research Task Description: This research project has been designed to focus on morphological developments during directional solidification of transparent analog hypermonotectic alloys. This is being accomplished by directionally solidifying transparent hypermonotectic samples using a temperature-gradient-stage assembly fitted to an optical microscope. Experimentation utilizes thin cells (20 μ m) oriented horizontally in order to minimize buoyancy driven convection and settling. This approach permits direct observation of the events at the solidification front. A range of growth rates, thermal gradients, and sample compositions have been utilized for two different systems, one with a high miscibility gap (succinonitrile-glycerol) and one with a low miscibility gap (succinonitrile-ethanol).

Progress to Date: Many interesting aspects of the solidification process in immiscible systems have been revealed in this investigation. For high-miscibility-gap samples, results indicate that it is possible to manipulate both the morphology and volume fraction of the immiscible phase through proper control of processing conditions. Perhaps one of the most significant findings is that the formation of the hypermonotectic phase can be suppressed during directional solidification so that L_2 does not nucleate in advance of the solidification front. This is accomplished through use of a high thermal-gradient to growth-velocity ratio in order to maintain a macroscopically-planar solidification front and steady-state growth conditions. Under these conditions, instead of the hypermonotectic phase forming in advance of the solidification front, the hypermonotectic phase participates in a cooperative growth process at the solidification front. The structure formed is identical to the monotectic structure (aligned and fibrous) with the exception that a higher volume fraction of the hypermonotectic phase is incorporated as aligned fibers.

Work in low-miscibility-gap samples has also led to many interesting observations. In low miscibility-gap samples, the cooperative growth process necessary for the formation of a regular aligned fibrous microstructure is not expected. However, our results indicate that a somewhat fibrous microstructure can be obtained at low solidification velocities. Unlike the high-miscibility-gap samples, there is a significant variation in the structure obtained with the fraction of the sample solidified. Work is still underway in this area.

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Microwave Materials Processing in Microgravity

Jet Propulsion Laboratory

Dr. Martin Barmatz

NAS7-918

October 1, 1981 - September 30, 1992

Objectives: The Microwave Materials Processing in Microgravity Task objective is to apply the unique capabilities of microwave heating and positioning to process the materials in microgravity. The task objectives will include 1) determination of the reaction mechanism, microstructure development and physical properties associated with microwave synthesis of ceramics, 2) development and application of microwave techniques for (a) monitoring the energy absorption during processing and (b) measuring thermophysical properties of materials in microgravity, and 3) theoretical modeling of unique microwave heating and positioning capabilities in a microgravity environment.

Research Task Description: There is a recognized need to produce advanced refractory ceramics that have higher melting temperatures and improved mechanical properties (such as strength and toughness). In recent years, ground-based experiments using microwave heating have demonstrated enhanced rates of sintering of ceramic materials leading to new microstructures. The synthesis of ceramics in a microgravity environment could provide the opportunity to produce contamination free ceramics with controlled microstructures that lead to advanced structural applications. Microwave processing can heat many glass and ceramic compositions very rapidly to high temperatures, it can heat them more uniformly than other methods, and it is energy efficient. Other important potential applications are crystal growth, and fiber pulling in space. Microwaves can generate a well defined temperature gradient within a material. This leads to the possibility of melting only the interior of a cylindrical sample, or leading to a radial gradient of the index of refraction upon solidification. By appropriate monitoring of the microwave parameters during processing one can also measure various sample properties as well as obtain energy absorption information which can be used to characterize the sample reaction and densification mechanisms. We have recently demonstrated that microwaves can produce unique positioning forces. The ability to microwave heat and position a sample may lead to a new containerless technology that is ideally suited for controlled processing of materials in microgravity..

Progress to Date: Drop shape analyses for the DPM triple axis positioner module were performed in preparation for a test of the theory during the USML-1 flight. Acoustic levitation studies at high ambient pressures, up to 20 atmospheres, demonstrated improved sample stability at higher ambient pressures as predicted by oscillational instability theories, and agreed with the enhanced force predictions of levitation theories. Successfully demonstrated controlled microwave heating of various materials in a cold walled cavity up to 1000 C.

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Dynamic Thermophysical Measurements in Microgravity

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Dr. Archie P. Miiller

W-17,428 (NASA Contact: R. Crouch, NASA HQ)

October 1, 1990 - September 30, 1991

Objectives: The main objective of this research is to develop accurate millisecond-resolution dynamic techniques which, in a microgravity environment, will enable the performance of thermophysical measurements on high-melting-point electrically-conducting materials in their liquid state at temperatures above 1500°K. The techniques when completed will enable, for the first time, the extension of accurate thermophysical measurements to temperatures above the limit (melting point) of the highly successful ground-based millisecond-resolution pulse-heating experiments. Thermophysical properties of interest include heat of fusion, heat capacity, electrical resistivity, thermal conductivity, surface tension, hemispherical total and normal spectral emissivities.

In addition to contributing to the advancement of general high-temperature science and technologies, the research is aimed at NASA flight support, specifically in the area of containerless processing. The research also has a strong component of shared technologies with other NASA supported programs in the areas of specimen heating, energy measurements, temperature mapping, and various other sophisticated high-temperature measurement problems.

Research Task Description: The research involves the development of a measurement system, employing millisecond-resolution dynamic heating techniques, for accurately determining selected thermophysical properties at high temperatures in a microgravity environment. The basic technique consists of resistively heating the specimen up to its melting point and above in about one second by passing a large current pulse through it, while simultaneously measuring the experimental quantities with millisecond resolution.

The research is comprised of two essential phases. The initial phase requires the establishment of criteria for the stability of specimens when heated rapidly to temperatures above their melting point in a microgravity environment. For this purpose, a test package has been designed and constructed to study the behavior of molten specimens under rapid heating conditions during microgravity simulations with NASA's KC-135 aircraft. The second phase of this work involves applying the basic technique to definitive measurements of selected thermophysical properties of one or more representative refractory metals (such as, niobium, tungsten, etc.) at and above their melting points.

As an integral part of the thermophysical measurements in microgravity, a millisecond-resolution multiwavelength pyrometer and a linear spatial scanning pyrometer are being developed to enable rapid and accurate measurements of the specimen temperature as well as temperature gradients in the specimen. The multiwavelength pyrometer (now fully operational) is capable of simultaneously measuring the radiance temperatures at six wavelengths (nominally in the range 500 - 900 μm) of a rapidly heating specimen every 0.5 ms. In addition to temperature measurements, this instrument will enable the study of other parameters relevant to optical pyrometry, such as optical properties (normal spectral emissivity, reflectivity, etc.) and easily-realizable high-temperature reference points. The linear spatial scanning pyrometer (development nearly complete) can measure spectral radiance temperature at about 1000 points along a straight line (25 mm long) on the specimen, with a complete cycle of measurements in about 1 ms. This pyrometer will be used to measure temperature gradients in a rapidly heating specimen providing data for diagnostic purposes and for determination of thermal conductivity, which will be a novel approach suitable for measurements at very high temperatures.

The overall system design will take into consideration the potential use of the system for other measurements and applications. Examples are: measurements of other thermophysical and related properties, measurements on other classes of substances, such as intermetallic compounds, investigations related to rapid solidification and supercooling, studies related to nucleation and kinetics of crystal growth, production and processing of pure materials, specialized fabrications, etc.

Progress to Date: The test package has been used, during several series of KC-135 flights, to study the behavior of molten specimens in various geometrical forms (solid rod, tubular, composite, triaxial, etc.), yielding valuable information regarding the parameters affecting specimen stability above the melting point under microgravity conditions.

A natural extension of the work on specimen stability with the test package has resulted in the development of a novel technique for measuring surface tension of liquid metals at high temperatures in a microgravity environment. The specimen geometry involves a tubular specimen mounted in a triaxial configuration in which a fraction of the heating current is returned along the tube axis. Adjustments to the current enable a balance between magnetic and surface tension forces acting on the molten specimen. Values for surface tension are determined from measurements of the equilibrium dimensions of the molten specimen tube, and the magnitudes of the currents. Rapid melting experiments, performed during the KC-135 flights, have yielded a value for the surface tension of copper at its melting point which is in good agreement with literature data. Preliminary microgravity experiments have also been performed on tantalum specimens in order to provide baseline data for future surface tension measurements. Theoretical/analytical studies of the effect of vibrations on the stability of liquid tubular specimens mounted in the triaxial configuration have also been carried out in support of the experimental work.

A microsecond-resolution technique has been used on ground to measure the heats of fusion of niobium, molybdenum, titanium and a titanium alloy, as well as the heat capacity and electrical resistivity of niobium near its melting point. These results will provide needed baseline data for performing significantly more accurate millisecond-resolution measurements under microgravity conditions in the future.

The multiwavelength pyrometer has been used to measure (on ground) the melting-point radiance temperatures (at six wavelengths in the range 522 - 906 μm) of several niobium strip specimens, yielding highly reproducible results to within about 1 degree kelvin. The high degree of reproducibility suggests that melting-point radiance temperatures of selected metals may provide the basis for high temperature references which can be easily realized for the rapid secondary calibration of optical pyrometers. This would be particularly attractive in environments where lengthy primary calibration procedures are either undesirable or not possible, such as in microgravity research at high-temperatures involving flight experiments (KC-135 aircraft, Space Shuttle or Space Station).

The development of a procedure for the automatic calibration of the linear spatial scanning pyrometer has been completed. Exploratory tests, involving the measurement of temperature profiles of rapidly heating specimens, have shown considerable promise for future applications of this instrument, not only in temperature diagnostics but also for determining thermal conductivity from measurements of axial temperature gradients.

Progress has been made in the design and construction of a new automated modular measurement system that will replace the existing test package. The measurement system includes a computer-based data acquisition and control module (CDACM), a high-speed camera module, an experiment chamber module, and a high-current power module. The CDACM has been assembled and ground-based testing is complete. The design of the other modules is underway. When complete, the new measurement system will provide the capability of automated in-flight diagnostics and data analysis, thereby making more efficient use of KC-135 flight time than is presently possible with the existing test

package. In addition, the modular design will provide greater flexibility in optimizing different requirements for measuring specific thermophysical properties of molten materials at high temperatures.

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The Dynamics of Disorder-Order Transitions in Hard Sphere Colloidal Dispersions

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April 1, 1990 - March 31, 1993

Objectives: These fundamental studies are aimed at a microscopic understanding of the most basic static and dynamic aspects of liquids, glasses, and solids and the solidification process. Our objectives are: (1) to complete the experimental characterization of the mechanics and dynamics associated with the disorder-order transition for hard spheres; (2) to ascertain the susceptibility of disordered systems to ordering by the application of external fields; and (3) to simulate the latter process via Brownian dynamics.

Research Task Description: This study will require a wide variety of experiments, preferably on a single well characterized, equilibrated, and unperturbed sample of a sterically stabilized silica suspension. Static, dynamic, and forced Rayleigh light scattering will be used, as well as viscometry to detect the structure and properties of lyophilic silica dispersions. Though laboratory experiments can be conducted in a novel fluidized bed apparatus, a microgravity environment will ultimately be essential to avoid the inhomogeneities introduced by settling during the slow phase transition and eliminate static density gradients and convection.

Progress to Date:

1. *Static Shear Modulus of Hard Sphere Sediments*

The group has developed a unique cell capable of measuring the shear modulus of sedimented crystals of sterically stabilized colloidal silica spheres. The usual problem associated with the measurement of the shear modulus measurement is that the particles typically sediment on the solid bottom of the container, so that the sediment is not accessible for displacement or force measurement. In order to overcome this problem, we let the particles sediment onto a liquid-liquid interface where the only shear restoring force comes from the colloidal crystal itself.

The colloids range from 0.1 to 1 micron in diameter and are soluble in cyclohexane. The shear cell consists of an octadecanol coated microscope cover glass floating on an interface between water and cyclohexane. The water is saturated with cesium chloride to ensure a greater density than the colloidal suspension. The colloidal crystal sediments on this interface. A small magnet on the bottom of the glass allows an external torque to be applied and a mirror attached to it allows for the reading of its angular displacement by means of a reflected laser beam.

Our measurements yield a shear modulus of several thousand dynes/cm². This value is orders of magnitude larger than the energy density or pressure in a gas of particles with the same density ($k_b T \times \rho$ the particle density). However, scaling the shear modulus with the pressure we expect that in equilibrium the shear modulus of the sediment should be directly proportional to the weight of the sediment above it. This yields the correct magnitude for the shear modulus and further provides an interesting way to study the equation of state of the hard sphere system. If one simply measures the density profile of the sediment in equilibrium, then the weight of the column above any height is just the pressure at that height.

We are presently measuring the shear modulus during the sedimentation process.

2. *World's Slowest Fluidized Beds*

In order to gain some insight into the properties of the hard sphere colloidal suspension in the microgravity environment, where a particular density can be stabilized in equilibrium (i.e., without a sediment), we have set up a fluidized bed. The idea is to keep the particles suspended against gravitational sedimentation by the viscous drag of the fluid. Since the particles of interest (those which most closely approach pure hard sphere interactions) settle at about 1 cm/day, this is the flow needed to prevent sedimentation, which makes this an extremely slow fluidized bed by any conventional standards.

In order to mimic the equilibrium conditions that we would like to attain in a microgravity environment, we have to assure that the state of the system is not strongly affected by the flow itself. The relevant parameter is the Péclet number on the scale of the interparticle separation. This is the ratio of the time for the particle to convect to a near neighbor compared to the time to diffuse the same distance. If the diffusion is much faster, then the system is effectively in equilibrium. The Péclet number is $0.2 (d/1\text{mm})^4$, where d is the particle diameter. Thus with our 0.5 mm particles, we are in the low Pe region and can study the equilibrium aspects of the liquid-solid hard sphere transition.

In a first set of experiments with the fluidized bed we have studied the sedimentation velocity of the silica spheres as a function of their volume fraction. This experiment shows the utility of the fluidized bed, in that the entire study of different concentrations was done with a single sample. Conventionally a sample is prepared with a known concentration and allowed to sediment. The speed of descent of the upper edge of the density profile provides a measure of the sedimentation velocity. Each concentration requires a separate sample. In the fluidized bed, the flow through the bed is set and measured, yielding the sedimentation velocity. The density is measured by optical techniques or by the steady state height of the bed. When the flow rate is lowered (raised), the density increases (decreases) to a new steady state value. The sedimentation velocity vs. concentration can thus be scanned continuously in a single sample. Similarly, using the fluidized bed we should be able in a single sample to continuously vary the density and study the other properties of the hard sphere system.

One of the main questions concerning the fluidization is whether the particle motion is due to thermal diffusion or to dispersion from the hydrodynamic interactions with the other particles in the flow. In order to study the relative contributions, we have to look on a length scale small compared to the interparticle spacing. At large distances both contributions look diffusive, while at small distances the hydrodynamic part looks convective (i.e., $\delta r(t) \sim t$) and can be distinguished from the diffusive part ($\delta r^2(t) \sim t$). The early time motion of colloidal particles is best probed with DWS, a technique which amplifies the individual particle motion by allowing the light to multiply scatter from the particles. We have set up a special fluidized bed with rectangular cross section and strongly scattering particles for DWS transmission experiments. Preliminary experiments show that both the convective and the diffusive short time behaviors can be measured.

3. *Transients in Slow Fluidized Beds*

The experiments exploring a range of concentrations with a given sample involve transients during which the height of the bed adjusts to a new flow rate. The slowness of the sedimentation process dictates rather long transients. Hence we must understand them in order to control or optimize the experiments and even extract information from the approach to steady state.

Analysis is straightforward, akin to the classical Kynch theory for transient settling. Thus the formation of a denser bed involves the propagation of discontinuities in concentration, with the top surface falling, a rising surface of the new fluid bed, and the potential for a fan of smoothly varying concentration between them. Its presence implies a "falling rate period" and a slower approach to steady state. Alternatively, expansion to a less dense bed occurs uniformly, at the convective velocity minus the settling velocity corresponding to the initial concentration.

The analysis suggests an interesting experiment with a crystal. First, the crystal must be formed through sedimentation at a low convective velocity to a high density. Second, the crystal is expanded progressively, in small steps, determining the settling velocity as a function of concentration and providing the opportunity to probe crystals of different densities. Ultimately the crystal will melt, yielding a disordered fluid just below the phase boundary. Third, a slight decrease in the rate of convection then causes the formation of a bed at a concentration across the phase boundary. Whether and how crystals form will depend on the rate of nucleation and growth relative to the rate of accumulation of the dense fluidized phase. Thus the series of measurements defines the settling velocity, the volume fraction at the disorder-order transition, and the dynamics of the transition.

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Modeling Directional Solidification Furnances/Processes

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In-House

Objectives: The underlying objective of this study is to develop a generic numerical tool for examining the interaction between the furnace and sample transport phenomena in directional solidification processes. A second goal is to implement an optimization procedure into the model to allow the modification of furnace components and/or operating parameters in order to achieve a user-designated design criterion (e.g. a prescribed solidification interface). The study will concentrate on flight furnaces and will provide a means for experimenters to model the interaction of specific hardware with their materials.

Research Task Description: A generic finite-element based code is chosen for computing the thermofluid transport phenomena in the furnace/sample. Specific enhancements to the code may include the following additions:

- heat of fusion using either a fixed grid (enthalpy) or moving grid approach.
- two solute transport equations.
- appropriate temperature and mass boundary conditions at the melt/solid interface.
- gray body radiation scheme
- furnace simplification algorithms (boundary curvature method).
- optimization loop to allow modification of user-selected regions (e.g. furnace components) and/or operating parameters as to optimize a designated criterion.
- database development for flight hardware.

The design tool will reside in a graphics workstation environment, complete with user interface and remote processing capabilities.

Space flight furnaces will be mathematically modeled to permit prediction of solidification behavior; more importantly, establishing predicted behavior will allow testing of the experimenter's understanding of the fluid physics involved in the process under study. The flight apparatus will be described in detail to allow realistic variation of process parameters. For experimenters with preferred growth rates, interface shape, and convective flow patterns, parameters will be adjustable for optimization.

Progress to Date: Improvements have been made to the Computational Materials Laboratory hardware and skills. As a trial program we have modeled at greater degrees of complexity the GaAs crystal growth experiment of GTE. A framework to accept the design details on the MSA and CGF furnaces has been constructed so that the designs and their modifications can be readily incorporated. In a related effort, the involvement of radiative heat transfer is being modeled so this important but frequently neglected mode of transport can be included. During the past year, design details have been placed into software. Predictions of growth rate and interface shape have been made for at least two of the accepted flight experiment with variation in the operational parameters considered. If problems are predicted, changes to the hardware which are within the range offered by the hardware contractors will be recommended. During the coming year, attention will be given to control of interface shaped through the application of shaped heating zones.

Current work under this funding has been directed to modeling multiple zone furnaces, modeling zone melting, and to solving the "inverse problem", e.g. given the desired interface shape thermal profile, and growth rate, what furnace settings are appropriate?

A complete model for float zone in an arbitrary thermal environment was formulated and coded for moderate surface-tension-driven convection.

A complete thermal model of laboratory prototype of the Programmable Multi Zone Furnace has been completed and verified. The model will enable rapid iterative thermal design of the next prototype, and in the future, available to all flight PI's for optimization of the growth parameters.

The Computational Material Laboratory at the MMSL was featured recently (October 1991) in the NASA Tech Brief as the cover and lead story. Many calls from industrial and academic interested parties were made following the story, and attempts will be made to deal with the growing demand for the lab.

Formulation of semi-transparent thermal radiation equations applicable to a wide range of optical and semiconductor materials has been done.

An interface code was developed to enable the interactive computations of transport processes during solidification together with the dislocation density map in the growing crystal.

Addition of magnetic field interaction with the flow field in conductive melts via the Lorentz force has been added in a general three dimensional formulation.

Addition of a general control module for simulating the dynamic behavior of furnaces for design application has been completed. The general purpose code can now be used both for thermal design problems (furnace/sample interaction) and for complete fluid, thermal, and solutal analysis of most solidification experiments both on earth and in space.

Demonstrated the Marangoni convection caused by different thermal environments using a float zone model, with emphasis on alternative furnace designs.

Publications

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Theory of Solidification

NASA Lewis Research Center

Professor Stephen H. Davis

NAG3-747

October 14, 1986 - October 9, 1992

Objectives: The research concerns the effort to understand on a quantitative level how various factors affect the morphology of a solidification front of binary materials. These factors include buoyancy-driven convection with and without Soret diffusion, forced flow, phase-change convection, crystal and kinetic anisotropies and effects of bounding surfaces. The central theme is the understanding of the phenomena through the study of the instability behavior of the appropriate coupled systems.

Research Task Description: The research entails the study of coupled systems using analytical methods, with the aim of identifying new mechanisms of behavior and new physical effects.

Progress to Date: In the past year we have concentrated on four aspects.

We have considered rapid solidification under conditions that the interface is governed by non-equilibrium thermodynamics. A self-consistent model is used and the morphological instability problem is reexamined. A pulsatile mode of instability is uncovered, whose interaction with the cellular mode, may explain the appearance of banded structures. The nonlinear behavior of this mode and its interactions have been examined as well.

We have examined Soret convection linked to a solid-liquid front. Both theory and experiment show how the coupling yield alterations of the interface morphologies.

We have considered the effects on interface stability of elastic stresses in the solid and surface diffusion. This work gives insight into the formation of islands in thin-film fabrication.

Two survey articles on the interaction of flow and solidification have been written. One has appeared in the Journal of Fluid Mechanics and one will appear in the Handbook of Crystal Growth. In addition Professor Davis is chairman of a NATO Advanced Workshop in March 1992, on the same topic.

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Convection in Partly Solidified Systems

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Professor Angus Hellawell
NAG-3-560

Objectives: The project is concerned with thermo-solutal convection during solidification of materials which freeze over a temperature range with a dendritic morphology. The research involves studies of metallic, aqueous and organic systems covering some three orders of magnitude in Prandtl number and is aimed at a quantitative comparison to demonstrate that channel convection is a generic phenomenon and that the dimensions, buoyancy forces and flow rates may be extrapolated.

Research Task Description: The experiments involve a base chill geometry: $\text{NH}_4\text{Cl-H}_2\text{O}$ and SCH-EtOH solidification is observed directly, Pb-Sn and Pb-Sb samples are examined after the events leading to channel segregation. Solute concentrations have been followed in convection plumes and channels and in bulk liquid and matrices. Plume temperatures have been measured in the transparent systems. In the aqueous material plume flow rates are around 10 mm s^{-1} , in the organic material around 1 mm s^{-1} . Analysis of these flow rates from measured quantities allows extrapolation to metallic systems in which it predicts flow rates approaching 0.2 m s^{-1} .

Progress to Date: During the past year, improved optical and video recording systems have greatly enhanced image resolution of convection patterns in the transparent materials and have produced more detailed and reliable data. The work has now been submitted for publication in some detail. Related studies of crystal movements and sedimentation rates have been made with the aqueous analogues and extrapolated to make predictive estimates concerning the columnar to equiaxed transition and equiaxed grain size in continuous steel castings.

Publications

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Jang, J. and Hellawell, A., "The Use of $\text{NH}_4\text{Cl-H}_2\text{O}$ Analogue Castings to Model Aspects of Continuous Casting, Part I, Asymmetry in Inclined Molds," *J. Ironmaking and Steelmaking* 18(4), 267-274 (1991).

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Containerless Processing of Oxide Superconductors

Vanderbilt University
Dr. William H. Hofmeister
Professor Robert J. Bayuzick
Dr. Richard Hopkins, Westinghouse R&D Center
NAG8-832 (NASA Contact: Marcus Vlasse, MSFC)

Objectives: Because of the high reactivity of oxide superconductors, they are an excellent candidate for containerless processing experiments. Benefits are expected from melt processing and undercooling of these materials that cannot be realized by conventional ceramic bulk processing techniques. The $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ or 1:2:3 compound has been chosen as a prototype for these studies for two primary reasons; the phase relations are reasonably well known, and as a peritectic compound, the possibility of solidifying the 1:2:3 phase from an undercooled melt is reasonably high. The experimental objectives are to explore the melting, undercooling and solidification behavior of these materials in a containerless environment and to characterize the microstructural and superconducting properties of the resultant materials. Comparison of these melt processed materials with ceramic, melt textured, and directionally solidified materials will help to elucidate the advantages or disadvantages of containerless processing on oxide superconductors. The techniques developed as a result of this project will be applicable to oxide superconductors in general and will provide additional avenues for research in this field.

Research Task Description: High purity powders from 10-300 μm of the 1:2:3 compound will be prepared by Westinghouse and processed in a 2 meter drop tube at Vanderbilt University. This drop tube has been specially designed to allow processing in oxidizing atmospheres and to provide for the melting and solidification of oxide superconductors. Full characterization of the resultant materials by both Vanderbilt and Westinghouse will be accomplished.

Progress to Date: The 2 meter drop tube equipped with a resistance heated muffle furnace has been assembled and tested with hot zone temperatures up to 1550°C. A dimpled quartz powder feed tube and a vibrating feeder have been implemented in order to control the rate of powder feed. Also, a pre-heating assembly has been installed to heat powders to 350°C prior to processing. The furnace has been equipped with two methods of temperature measurement: a W, 5%Re - W, 26%Re thermocouple connected to a chart recorder and a single color brightness pyrometer with access to the element via a viewing port. In addition, the assembly allows for processing of powders in a controlled atmospheric gas mixtures, free from container contamination. Powders from 50-200 μm of the 1:2:3 compound have been successfully processed in a 20% oxygen-80% helium gas mixture. The resultant spheres are 50-200 μm in diameter. Analysis shows that these spheres consist of cubic Y_2O_3 , tetragonal $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$, and a mixture of BaCu oxides. The yttria exists as faceted precipitates in a mixture of the other compounds. This suggests that a yttria plus liquid exists at high temperatures at the 1:2:3 composition, and that full melting has not occurred. Annealing the spheres first at 900°C and then at 400°C in flowing oxygen results in single phase orthorhombic 1:2:3 at the starting composition.

Steps are under way to increase the maximum temperature achievable in the furnace in order to fully melt the yttria prior to solidification. Exchanging the alumina muffle tube with a zirconia muffle tube will allow for maximum furnace temperatures of approximately 1900°C. TEM analysis and superconducting property measurements are in progress.

Modeling and Experimental Studies of Droplet Pushing in Miscibility-Gap Alloy Solidification Under Low-G Conditions

University of Colorado
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D. B. Thiessen
NAG3-1278
June 1, 1991- October 1, 1991

Objective: The processing of metallic miscibility-gap alloys in a low-gravity environment has the potential to produce advanced materials with unique microstructures. Materials produced in this way would have applications as superconductors or in optoelectronic devices. Previous low-gravity processing studies have not succeeded in producing the desired microstructures. This research seeks to explore one of the important mechanisms governing microstructure development in the processing of miscibility-gap materials under low-gravity conditions. In particular we wish to understand the mechanism of droplet pushing by a growing solidification front. The first objective of the research is to obtain qualitative and quantitative information on the droplet-pushing mechanism by observation of the solidification front in transparent organic miscibility-gap systems. The second objective is to develop a mathematical model of droplet pushing which predicts the critical drop diameter as a function of processing conditions and fluid properties. The final objective is to determine the proper form of the thin film model for the droplet-pushing problem. The results of this research will be used to establish the proper processing parameters in order to obtain the desired microstructures in miscibility-gap alloys.

Research Task Description: Preliminary work on the study of the droplet pushing problem was supported by the Center for Low Gravity Fluid Mechanics and Transport Phenomena at the University of Colorado (NASA Grant # NAGW-951). The current work on this project builds on this preliminary work; therefore, a brief description of this preliminary work will be given. An integrated program of experimental and modeling work was proposed. One of the investigators (DBT) spent June-September 1989 at NASA-MSFC working with Dr. Donald O. Frazier on experimental solidification studies of transparent organic miscibility-gap alloys. These studies utilized NASA's gradient-stage microscope facility. The results of these experimental studies confirmed that droplet pushing could occur in a miscibility-gap system and elucidated important effects to be considered in the modeling work. Critical droplet-size measurements were made as a function of solidification rate in the succinonitrile-camphene system. Following this, a model of droplet pushing was developed which accounted for the mobile interface of the droplet. This model predicted critical drop size as a function of solidification rate. Comparison of the model results with the experimental measurements was not possible without knowing the Hamaker constant of the organic materials studied. A new technique for obtaining the Hamaker constant from interfacial tension data on the system was developed, although the necessary experimental interfacial tension data were not yet available.

Progress to Date: Since the commencement of the current grant, efforts have been directed toward measuring the interfacial tensions of the succinonitrile-camphene system in order to make quantitative comparisons between model and experiment. A zone-refining apparatus has been constructed in order to purify the organic materials. A pendant drop apparatus for measurement of interfacial tension has been set up and is currently being tested. The mathematical model is being extended to include solute field effects on the local growth rate of the solid in the vicinity of a growing droplet.

Publications

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Presentations

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Containerless Processing of Undercooled Melts

University of Wisconsin-Madison
Professor John H. Perepezko
Grant NAG-771 (M.B. Robinson MSFC)
October 1990 - October 1991

Objective: The main objective of the research project is to evaluate the undercooling and resultant solidification microstructures in containerless drop tube processing of the alloys as an experience base for microgravity experiments.

Research Task Description: The degree of liquid undercooling attainable in a laboratory scale (3m) drop tube can be altered through the variation of processing parameters such as melt superheat, droplet size and gas environment. In a given sample, the competitive nucleation and growth kinetics between equilibrium and metastable phases controls microstructural development. The solidification behavior is evaluated through metallography, thermal analysis and x-ray diffraction examination in conjunction with calorimetric measurements of falling droplet temperatures and a heat flow model of the processing conditions to judge the sample thermal history.

Progress to Date: In the current program studies, solidification microstructures are being examined in selected Ni, Fe, and Mn based systems. The specific alloy selection is based on a metastable phase diagram analysis that allows for the identification of unique microstructures and microstructural transitions that may be produced by microgravity containerless processing. For example, with this approach the apparent dissimilarities reported for the solidification behavior in undercooled Fe-Ni alloys as a function of sample size and processing conditions have been resolved by our studies on samples covering seven orders of magnitude in volume. From this analysis a processing map of solidification pathways and microstructures has been developed that offers a clear and consistent delineation between powder and bulk sample behavior important for sample scale-up for microgravity experiments. In a similar manner, a duplex partitionless solidification reaction has been identified in several alloys near the eutectic composition in the Ni-V system. The reaction can be thought of as a limiting case of a eutectic process, $L \rightarrow \alpha + \beta$ in which α and β have the same composition as the liquid. Drop tube experiments are being used to understand and model the competitive kinetics controlling the duplex partitionless reaction. Near equiatomic Mn-Al alloys represent an important class of permanent magnet materials. The key ferromagnetic phase is a metastable structure produced by solid state heat treatments. Recent drop tube studies have demonstrated for the first time that the metastable ferromagnetic phase can be produced from the liquid provided high undercooling is achieved. With specially prepared samples it has been possible to assess the thermodynamic driving forces involved in metastable phase solidification. Building on the thermodynamic analysis a competitive nucleation model is being developed to account for the observed phase selection. As part of a test of the kinetics model a detailed statistical study of the metastable product yield as a function of sample size and processing gas is underway. In the above studies a new calorimetric system is being used to measure the temperature of falling drops during containerless processing. In another study on drop tube processing of magnesium metasilicate liquids, a glass-ceramic microstructure has been produced in samples that undercool into a liquid phase miscibility gap before the onset of crystallization. A kinetic study of the competition between vitrification and crystallization in this system is in progress.

Publications

Cooper, R.F., Yoon, W.Y., and Perepezko, J.H., "Internal Nucleation of Magnesium Metasilicate Glasses by Solidification Processing," J. Am. Ceram. Soc. **74**, 1312 (1991).

Thoma, D.J. and Perepezko, J.H.m, "Microstructural Transitions during Containerless Processing of Undercooled Fe-Ni Alloys," Met. Trans., 1991 (accepted).

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Thermosolutal Convection and Macrosegregation in Dendritic Alloys

University of Arizona
Professor D. R. Poirier
Professor J. C. Heinrich
NAG 3-1060 (NASA Contact: T. K. Glasgow, LeRC)
July 18, 1989 - November 27, 1991

Objectives: The major objective of the program is to develop a computer model for studying thermosolutal convection and associated macrosegregation phenomena in dendritically solidifying alloys.

Research Task Description: Comparison between the calculated macrosegregation and experimental results of other investigators will be made. A major feature of the code will be to simulate the nonlinear convection that leads to the formation of a macrosegregation defect, called freckles. The calculations will include the effects of varying solidification rate, thermal gradient, and composition in Pb-Sn and Pb-Sb alloys. Time permitting, comparisons between calculated and experimental results in solidified $\text{NH}_4\text{Cl-H}_2\text{O}$ solutions will also be made.

The effects of reducing the magnitude of gravitational acceleration and changing its direction on thermosolutal convection and the attendant macrosegregation, will be studied by using our finite element code for the fully nonlinear convection problem. The goal of these simulations will be to assist in planning experiments in low-gravity environments and to elucidate the thermosolutal convection that occurs in dendritically solidified alloys.

Progress to Date: The finite element code (used for grant NAG3-723) has been modified so that it can be used to simulate unsteady dendritic solidification in a vertical mold. An important feature of the code is that it calculates the adjustment of the volume fraction of the interdendritic liquid, as convection occurs in the mushy zone. Calculations have been done for a Pb-Sn alloy which show the development of freckles during directional solidification--the first of their kind. In addition, pockets of segregated liquid form within the mushy zone, itself, and the all-liquid zone selectively penetrates into the mushy zone producing additional segregation zones.

Publications

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Heinrich, J. C., Felicelli, S. D., and Poirier, D. R., "Freckles in Directional Solidification of Binary Alloys," MECOM 91, *Proceedings of Latin American Congress on Computational Mechanics*, Santa Fe, Argentina, September 23-25, 1991, to be published.

Containerless Studies of Nucleation and Undercooling

Jet Propulsion Laboratory
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Dr. Kenichi Ohsaka, Cal Tech
January 1991 to January 1992

Objectives: The long term research objectives are experimental and theoretical studies to determine the achievable limits of undercooling using ultrasonic and other means of sample levitation and manipulation, to study the characteristics of heterogeneous nucleation using levitated materials in 1-g and in microgravity, to measure the physical properties of significantly undercooled melts, and to determine the characteristics of solidification under conditions allowed by both containerless processing and low gravity.

Research Task Description: Ground-based experiments and investigations aboard the NASA KC-135 aircraft are carried out to levitate, melt, undercool, and solidify 0.5 to 3 mm specimens of low melting pure metals and alloys (Ga, In, Sn, AL, Al-Cu alloys, Ti-Cr alloys, and glass-forming alloys), as well as organic compounds (Ortho-Terphenyl, Succinonitrile), and low melting glasses. Non-perturbing techniques for the measurement of the density, surface tension, viscosity, refraction index, sound velocity, specific heat, and thermal diffusivity are being refined and developed to probe the physical state of levitated undercooled melts. The quantitative evaluation of the effects of external physical stimuli on the nucleation onset is also being carried out to rigorously document the advantages of experimentation in microgravity. In addition to the undercooled liquid phase, the solidification process under containerless conditions has also been investigated for model materials in 1-g. Closer examination of the thermal and flow environment within and outside the melt is being carried out experimentally and theoretically. Current experimental approaches involve the utilization of controlled-environment ultrasonic and electromagnetic levitators and noncontact probing techniques..

Progress to Date: 1-g undercooling of a wide variety of materials has demonstrated that the containerless environment indeed enhances the ability to access the metastable state. In addition, definite evidence that 1 g-induced secondary sample perturbations due to the levitation mechanisms are drastically reduced in low gravity has been obtained using ultrasonic devices and KC-135 experiments. The undercooling and dendritic solidification of levitated Succinonitrile has been carried out, and preliminary measurement of the dendritic growth velocity on the surface of a spheroidal droplet has been obtained. Experimental evidence of ultrasonically-induced nucleation of undercooled succinonitrile has also been gathered. The capability for hypercooling ultrasonically levitated Succinonitrile bulk samples has also been demonstrated. The undercooling of electromagnetically levitated Aluminum and Titanium -Chromium alloys has been performed together with the measurement of the specific heat using a drop calorimeter. Both theoretical and experimental work are being carried out to develop remote techniques for the measurement of the thermal diffusivity of levitated melts in 1-g.

Publications

Trinh, E.H., "Fluid Dynamics and Solidification of Drops and Shells," in *Progress in Low Gravity Fluid Dynamics and Transport Phenomena*, Volume 130 (J. Koster and R. Sani, eds.), AIAA, 1990, p. 515.

Ohsaka, K., Gatewood, J., and Trinh, E.H., "An Apparatus for the Specific Heat Measurement of Undercooled Liquids," *Scripta Met. et Mat.* 25, 1459 (1991).

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Ostwald Ripening of Solid-Liquid Mixtures

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Objectives: The objective of this program is to use the unique conditions provided by spaceflight to study the kinetics of Ostwald ripening. The data derived from this experimental work will provide baseline data for the field and thus permit the refinement of existing theories of the kinetics of first-order phase transformations. In addition, as the Ostwald ripening process has a major impact on the properties of materials, the experimental results will yield information which can be used to improve the properties of materials containing dispersed phases, such as precipitation hardened alloys.

Research Task Description: An ideal system to use in these experiments is a two-phase mixture consisting of solid particles in a liquid. Since the coarsening rate of the solid particles in such a system is comparatively fast, and in a properly chosen system the particles can be spherical, the experiments can serve as a careful test of theory. However, experiments performed in systems with a low volume fraction of solid particles, under terrestrial conditions shows that buoyancy driven convection of the solid particles is prevalent. Thus the experiments do not satisfy the theoretical requirements of fixed spatial locations of the particles. To eliminate this problem, it is necessary to perform the experiments in the reduced gravity environment of space.

Progress to Date: Experimental measurements of isothermal grain boundary grooving by volume diffusion were carried out for Sn bicrystals in the Pb-Sn system near the eutectic temperature. The dimensions of the groove increase with a temporal exponent of $1/3$, and the measurement of the associated rate constant allows the determination of the product of the liquid diffusion coefficient and the capillary length associated with the interfacial free energy of the crystal-melt interface. We generalized the small-slope theory of Mullins to the entire range of dihedral angles by using a boundary integral formulation of the associated free-boundary problem, and obtain excellent agreement with the experimental groove shapes. By using the diffusivity measured by Jordan and Hunt, we deduce that our measured values of the capillary length agree to within 5% with the values obtained from experiments by Gunduz and Hunt on grain boundary grooving in a temperature gradient.

In an effort to understand the dynamics of Ostwald ripening in systems consisting of a high volume fraction array of spherical solid particles in a liquid, we have developed an approach which allows the diffusion field in the liquid matrix to be modeled to an arbitrary degree of accuracy. This is done by writing a solution to the diffusion equation in terms of integral equations and then solving these integral equations in terms of spherical harmonics. To date, in addition to monopoles, we have included dipolar and quadrupolar interactions between the particles. We have found that these additional terms have a large effect on the coarsening rate of particles for volume fractions above approximately 0.1.

The Lifshitz-Slyozov (LS) theory for coarsening predicts that the average particle size grows as $t^{1/3}$ and that a self-similar attractor-state particle size distribution (PSD) exists. Both predictions are strictly valid only in the limit of infinitely long times and zero volume fraction of coarsening phase. In characterizing transient coarsening behavior, the time-evolution of selected initial PSD's have been studied via a numerical solution of the continuity equation. Initial PSD's included gaussian and log-normal forms, neither of which reached the self-similar attractor state by dimensionless time $t = 10^3$, corresponding to nearly an order of magnitude change in the average particle radius. Yet, from scaled plots of the PSD's and from phase-plane plots of the moments of the PSD's, the observed behavior was consistent with that expected for a system approaching the LS attractor-state. The effect of a finite

volume fraction of coarsening phase on the dynamics of transient Ostwald ripening has also been investigated.

Publications

Akaiwa, N. and Voorhees, P.W., "The Effects of a High Volume Fraction of Coarsening Phase on the Dynamics of Ripening in Solid-Liquid Mixtures," in *Proceedings of the Conference on the Nature and Properties of Semi-Solid Materials* (J.A. Sekar, ed.), 1991.

Hardy, S.C., McFadden, G.B., Coriell, S.R., Voorhees, P.W., and Sekerka, R.F., "Measurement and Analysis of Grain Boundary Grooving by Volume Diffusion," J. Cryst. Growth, 1991 (in press).

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Influence of Convection on Microstructure

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Dr. Rubens Caram
Ms. Jayshree Seth
Mr. James Rydzewski
NAG8-753 (NASA Contact: Dr. F. Szofran, MFSC)
August 1988 - August 1992

Objective: The objective is to gain an understanding of the influence of microgravity on the microstructure of eutectics, especially MnBi-Bi.

Research Task Description:

1. Via theoretical calculations, determine the influence of convection on the microstructure of fibrous eutectics.
2. Via theoretical calculations, determine the influence of the Soret effect (thermal diffusion) on eutectic microstructure.
3. Estimate the Soret coefficient of eutectic MnBi-Bi melts using laboratory data and data from the flight experiment of Dr. David Larson at Grumman.
4. Via theoretical calculations, determine the influence of convection on eutectic microstructure when one phase projects out into the melt ahead of the other phase.
5. Experimentally determine the influence of vibration during solidification on eutectic microstructure.
6. Experimentally determine the influence of centrifugation during solidification on eutectic microstructure.
7. Experimentally determine the influence of deviations from eutectic composition on the microstructure of directionally solidified MnBi-Bi.

Progress to Date: Tasks 1, 2, 4 and 5 above were completed. Task 3 was partially completed. Convection is predicted to coarsen eutectic microstructure more when the structure is rod-like rather than lamellar, and more when one phase projects out than for a planar interface. However the effect is still not large enough to explain the experiments of Larson and Pirich that showed a two-fold decrease of MnBi rod spacing when solidification was carried out in space or with a magnetic field applied.

Our theoretical results predict that the Soret effect can either decrease or increase the inter-phase distances in eutectic solidification, depending on the sign of the Soret coefficient. Again it does not seem likely that this effect is large enough to explain the Larson/Pirich results.

Our ground-based experiments designed to yield values of the Soret coefficient for Mn-Bi eutectic were interesting but inconclusive. The eutectic was placed in capillary tubes and held vertically in a furnace with the temperature increasing with height. A rapid separation occurred. This separation was attributed to settling of the slow-melting phase during heatup. After many days, steady state concentration profiles were obtained that suggest the presence of very gentle buoyancy-driven

convection. Flight experiments should be designed aimed specifically at measuring the Soret coefficient, because buoyancy-driven convection appears to be impossible to completely avoid on earth.

Vibration during solidification of lead-tin eutectic produced a large increase in the interlamellar spacing. This increase is beyond that expected for the amount of convection produced. Our previous experiments with spin-up/spin-down (accelerated crucible rotation) did not change the lamellar spacing at all. On the other hand, vibration gave no separation of lead and tin, while a significant separation was obtained with spin-up/spin-down. Spiralling of the microstructure as observed with vibration did not occur.

A numerical model predicted that convection has a larger influence on lamellar spacing when the interface is stepped than when it is planar, i.e. when one phase projects out ahead of the other. Again the predicted effect is not sufficiently large to explain the flight results of Larson and Pirich.

The MnBi-Bi eutectic was directionally solidified in a large centrifuge in the USSR. The microstructure is being determined. We prepared ampoules containing MnBi-Bi eutectic and off-eutectic for solidification here and in centrifuges in Canada and the USSR.

Publications

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3. FLUIDS, INTERFACES, AND TRANSPORT

Residual Acceleration Data on IML-1: Determination of Experiment Sensitivity and Efficient Data Dissemination

University of Alabama in Huntsville
Center for Microgravity and Materials Research
Dr. J. Iwan D. Alexander
NAG8-759 (NASA Contact: Dr. R. S. Snyder, MSFC)
March 1989-February 1992

Objective: The objective of this research is to develop a mission-specific residual acceleration data reduction and dissemination plan. It is designed to meet the needs of individual experimenters and result in a useful, manageable data base. This will enable the principal investigators to assess the acceleration environment to which their experiment was exposed, without having to examine the entire acceleration history recorded during the experiment (400 Mbyte/disk, some 20 disks/mission).

Research Task Description:

1. Identification of sensitive experiments and sensitivity ranges (magnitude and frequency) through order of magnitude estimates and the results of numerical modelling.
2. Research and development toward a plan for reduction and dissemination of residual acceleration data.
3. Use of a "Shuttle Motion Model" to predict accelerations arising from specific Shuttle maneuvers.
4. Supplementation of recorded data with calculated predictions for gravity gradient and other accelerations resulting from the basic orbital motion.
5. Implementation on existing acceleration data bases.

Progress to Date: Several specific experiments and general experiment classes have been identified as potentially sensitive to perturbations of the low-gravity environment in an orbiting laboratory:

- o Critical point experiment (CPF)
- o Crystal growth from solution (TGS/FES)
- o Growth of mercuric iodide from the vapor (VCGS/MICG)
- o Casting and solidification technology (CAST)
- o Protein crystal growth. (PCG)
- o Gravitational plant physiology experiment (GPPF)
- o Organic crystal growth experiments (OCGF).

Results from order of magnitude analyses and numerical simulations indicate that low frequency and steady accelerations may be of concern.

The research and development aspects of the data reduction plan have progressed and a prototype plan was implemented on the Spacelab 3 (SL3) accelerometer data base. Discussions of the basic analysis techniques used and the results achieved to date are included in some of the publications. In addition, we have attended and presented summaries of our processing techniques and results at several Microgravity Measurement Group (MGMG) and IML-1 IWG meetings.

The two-level data reduction plan developed will enable principal investigators to create a limited, user-specific accelerometer data base that can be easily merged into the post-flight experiment analysis process. In addition, the processed data, particularly that associated with specific crew activities, will contribute to the characterization of the acceleration environment of the Orbiter. Comparison of

windows of the SL3 data with data collected on other orbiter missions has already led to the identification of acceleration signatures related to specific structures and pieces of equipment in the orbiter Columbia. With the flight of IML-1, the acceleration environment characterization process can be expanded to the orbiter Atlantis.

In general, three main features of residual acceleration data can be used to characterize the acceleration environment. These features are the *time history* of the data (maximum values, mean values, acceleration vector magnitude, and particular disturbance patterns), the *frequency* components present in a given window of data, and the *orientation* of the acceleration vector during that time window. These features of residual acceleration data also play an important role in the creation of user-specific databases and in the post-flight processing of experimental results in conjunction with accelerometer data.

The amount of data processed and the form in which it will be most useful to individual PIs will depend on the particular experiment. The first level of our two-level data reduction plan identifies factors specific to an experiment that will be important in the evaluation of the needs of the investigator. The second level of the data reduction plan involves actual data processing based on the results of the first stage of the plan. The processing done at this level will result in limited, user-specific accelerometer data bases that principal investigators can use in the post-flight analysis of their experimental results.

The current phase of our work leading to the IML-1 mission in 1992, involves the identification of specific processing techniques that will be useful for principal investigators in the post-flight analysis of experimental results in conjunction with accelerometer data. A peak detection routine has been used (on large data segments from SL3) to identify acceleration events that have exceeded specified acceleration tolerances. These data are then analysed by various techniques. In particular, we have focused on data decimation, cluster analysis techniques and various 3D data representations to enable the investigation of low frequency information from long time histories and on cross-correlation analysis to identify causal relationships between accelerations and experiment perturbations.

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Thermo-Diffuso Capillary Phenomena

NASA Lewis Research Center
Dr. R. Balasubramaniam
Dr. L. H. Dill
In-House
Continuing Task

Objectives: The main objective of this program is to understand the motions of bubbles and droplets due to thermocapillary effects (Marangoni flow) in a reduced gravity environment.

Research Task Description: Analytical and numerical studies are being performed to predict the terminal and transient velocities of immiscible bubbles and droplets in a host fluid possessing a uniform temperature gradient. The terminal velocity of a bubble has been calculated including the effects of inertia and convection, i.e., for non-negligible values of the Reynolds and Marangoni numbers. The steady thermocapillary migration of a gas bubble in the presence of an insoluble surfactant is also being analyzed.

Experiments are also being conducted using an immiscible density matched liquid-liquid system to obtain migration data for droplets and also visualize the thermocapillary flow that is induced within the droplet.

Progress to Date: In the past year, experiments have been performed successfully on visualization of thermocapillary flows in drops of vegetable oil, water-methanol mixtures and pure methanol in a host liquid of silicone oil. Attempts were made to visualize the temperature field using interferometry in test cells filled with silicone oil. Theoretical analyses have been completed predicting the thermocapillary migration velocity of a compound drop and the results were adapted to a thin-shell limit. An Oseen-like approximation was used in the energy equation to analyze the migration velocity behavior, thermal boundary layer and wake structure for a migrating bubble. Attempts were also made to investigate the thermal boundary layer structure with a realistic flow field. Thermodynamic analyses have been completed showing the dependence of surface tension on electric charge and a manuscript for publication is in preparation. Significant effort was devoted to defining flight experiments for conduct in the Bubble, Drop, and Particle Unit (BDPU). Assistance was provided to the flight experiment principal investigator to prepare a Science Requirements Document, address technical issues regarding thermal relaxation times, refine the experiment test matrix, prepare for an investigators working group meeting and provide comments on many letters and fax communications with the hardware manufacturers.

Publications

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Dill, L.H. and Balasubramaniam, R., "Unsteady Thermocapillary Migration of Isolated Drops in Viscous Flows," Int. J. Fluid Flow, 1991 (in press).

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Fluid Mechanics of Directional Solidification at Reduced Gravity

University of Arizona
Professor C. F. Chen

Objectives: The primary objective of the proposed research is to provide additional ground-based support for the flight experiment "Casting and Solidification Technology" (CAST), with principal investigators Drs. M. H. McCay and T. D. McCay of the University of Tennessee Space Institute, to be flown in a space shuttle mission scheduled for 1992. In particular, we will provide data on the convective motion and freckle formation during directional solidification of nitrogen hydrogen chloride (NHCl) from its aqueous solution at simulated parameter ranges equivalent to reducing the gravity from the sea-level value down to 0.1 g and lower.

The secondary objectives of the proposed research are to examine the stability phenomena associated with the onset of freckles and the mechanisms for their subsequent growth and decline (to eventual demise of some) by state-of-the-art experimental techniques and to formulate mathematical models for the prediction of the observed phenomena. The novel experimental technique to be used is computed tomography, which is capable of presenting a three-dimensional image reconstruction of the entire mushy zone. This will enable us to observe the initiation of the chimneys in the mush and their subsequent evolution. It will also enable us to determine quantitatively the variation of porosity within the mushy zone with a spatial resolution of $0.25 \times 0.25 \times 2$ mm. The experimental insight gained will provide us with the necessary physical background to construct a rational model for the prediction of such a complex process.

Research Task Description: This project involves both experimental and theoretical aspects. Three sets of experiments on the solidification of aqueous solutions of NHCl are planned. The first set will be conducted in order to determine the effect of reduced gravity and the critical conditions for the onset of plumes. The second set is designed to be carried out with computed tomography. The third set will be run at high initial temperature gradients, $\sim 8^\circ\text{C}/\text{cm}$, the same as those used by McCay, McCay, Lowry, and Smith (1988) in their ground-based experiments. The results of the first and third sets will provide ground-based support for the CAST experiment in space, and the second set of experiments will exploit the technique of computed tomography in order to determine the characteristics of convection in the mushy zone.

In the theoretical aspect, we plan to construct a model of the convection process occurring in the mushy zone for the prediction of the onset of plume convection. We also plan to perform nonlinear computation using initial and boundary conditions similar to the ones used in the experimental investigation for the purpose of comparing the two sets of results. In particular, the calculated porosity distribution in the mushy zone will be compared to the values obtained by the CT scan.

Progress to Date: The grant was awarded on April 1, 1991. Progress made on this project in the past six months is as follows:

The paper "Experimental Study of Directional Solidification of Aqueous Ammonium Chloride Solution" by C. F. Chen and Falin Chen has appeared in the Journal of Fluid Mechanics (1991, 227, 567-586). This work was carried out under the previous grant, NAG 3-723. Included in this paper are results of onset of plume convection from the mushy zone down to an equivalent gravity level of 0.1 g, where g is the gravity at sea level. The results also show that salt-finger convection above the mushy zone will persist to a gravity level of 10 g. Experimental apparatus has been designed and is being constructed for carrying out experiments with computed tomography. Discussion is underway with medical staff at the CT scan laboratory to find ways to scan the growing mushy zone while the solidification process is under way. The effect of g-jitter on the onset of instabilities in a double-diffusive layer with cross-diffusion has

been analyzed using linear stability analysis with Floquet theory. Results show fundamentally different features in the topology of neutral curves and stability boundaries not found in singly diffusive layers under g-jitter. Among the more important findings are: (1) The existence of neutrally stable periodic solutions that are neither synchronous nor subharmonic. (2) Neutral curves exhibiting multiple bifurcation points connecting three different classes of neutral solutions; (3) The existence of two onset incommensurate frequencies at two different wave numbers for the same Rayleigh number. This work is part of the Ph.D. dissertation of Mr. Guillermo Terrones. Dr. Chen is his thesis advisor. Mr. Terrones will present a summary of his results at the American Physical Society's Division of Fluid Dynamics Annual Meeting in November 1991 and a full paper version at the AIAA Aerospace Sciences Meeting in January 1992. The effect of surface tension on the onset of instability in a double-diffusive layer has been analyzed using linear stability theory. Among many interesting results is the fact that salt-finger instability, which onsets into steady convection under normal conditions, onsets in oscillatory mode under a range of parameters. A full paper version of this work will be presented at the AIAA Aerospace Sciences Meeting in January 1992. During the summer, June 1-August 5, 1991, Dr. Chen was a Visiting Fellow at the Research School of Earth Sciences, Australian National University. He carried out experiments on directional solidification of NHCl in the Geophysical Fluid Dynamics Laboratory of Professor J. S. Turner. Using flow visualization techniques and time-lapse motion pictures, he was able to record the convective motion in the mushy zone before and after the onset of plume convection. He also carried out directional solidification experiments in a Hele-Shaw cell. The evolution of chimneys was clearly exhibited and recorded by close-up photography with 1:1 magnification. Dr. Chen presented a summary of these results at the APS Division of Fluid Dynamics Meeting in November 1991.

Publications

Chen, C. F., "Surface Tension Effects on the Onset of Double-Diffusive Convection," presented at the IUTAM Symposium on Microgravity Fluid Mechanics, Bremen, Germany, September 2-6, 1991.

Chen, C. F., "Directional Solidification of NHCl Solution in a Hele-Shaw Cell," to be presented at the 44th Annual Meeting of the Division of Fluid Dynamics of the American Physical Society, Tempe, Arizona, November 1991.

Chen, C. F. and Falin Chen, "Onset of Salt-Finger Convection in a Gravity-Gradient," Physics of Fluids, 1991 (submitted).

Chen, C. F. and T. F. Su, "Stability Analysis of Surface Tension Effects on a Double-Diffusive Layer," AIAA paper to be presented at the AIAA 30th Aerospace Sciences Meeting, January 6-9, 1992.

Terrones, G., "Gravity Modulation and Cross-Diffusion Effects on the Onset of Multicomponent Convection," Ph.D. dissertation, Dept. Aero. and Mech. Eng., The University of Arizona, November 1991.

Terrones, G. and C. F. Chen, "G-Jitter and Soret Effect on the Onset of Convection in a Doubly Diffusive System," AIAA paper to be presented at the AIAA 30th Aerospace Sciences Meeting, January 6-9, 1992.

Fluid Interface Behavior under Low- and Zero-Gravity Conditions

University of California, Berkeley
Professor Paul Concus
Professor Robert Finn (Stanford University)
NAG3-1143

Objectives: The general objective of this research is to gain better mathematical understanding of the physical behavior of fluids partly filling a container or otherwise in contact with solid support surfaces, when capillary forces predominate. Closely interrelated with the mathematical and computational studies are current and planned ground-based and in-space microgravity experiments.

Research Task Description: Work currently is directed (a) toward the realization of experiments designed to observe actual behavior of equilibrium fluid configurations in "exotic" containers for which exact theory predicts an entire continuum of symmetric surface interfaces, but for which it can be shown that no symmetric interface can be stable. Our second (and independent) direction (b) is toward the design of experiments for very precise measurement of contact angle; these latter experiments are intended also as a test of the physical validity of the concept of contact angle, as envisioned by the classical Young-Gauss theory.

The specific design and construction of the experimental apparatus for item (a), scheduled for flight on USML-1, is being carried out by M. Weislogel at NASA Lewis Research Center and for item (b), which is proposed for flight under the aegis of the European Space Agency, is being carried out in collaboration with D. Langbein, U. Hornung, M. Haynes, and M. Weislogel. In both cases our responsibility rests primarily with the mathematical and computational theoretical underpinnings.

Progress to Date: A systematic numerical calculation of the exotic containers has been completed over a range of parameters of physical interest. Preliminary drop-tower experiments have been carried out by M. Weislogel at the NASA Lewis Zero Gravity Facility, and in some of the experiments the fluid reoriented from its initial symmetric configuration during the period of free fall to one that was not symmetric, in corroboration of the mathematical theory. The time scale was too short to obtain scientifically definitive results, but the experiments provided needed information for the design of longer-duration microgravity experiments in space. The design for the USML-1 experiments has been essentially completed, as well as construction of the experimental apparatus. Numerical calculations to seek energy-minimizing fluid configurations in the exotic containers were carried out in collaboration with M. Callahan, and of the several distinct configurations obtained the one having least energy looks remarkably similar to the surface indicated in the drop-tower experiments.

In connection with item (a) above, joint work with T.I. Vogel on liquid bridges between parallel plates was carried out. We have proved that for any contact angle the greatest lower bound on fluid volumes at which such a bridge can be stable always exceeds the value that occurs for contact angle 90 deg. Since in this latter case the critical volume is known explicitly, we obtain a working estimate valid for any contact angle. In this way are able to design exotic containers so as to exclude the occurrence of (unwanted) liquid bridges between the top and bottom surfaces. The investigation is continuing with a view to characterizing what happens when the contact angles on the two surfaces differ.

With regard to the forthcoming space experiments, an estimate was required of the critical contact angle for a cylindrical container with rectangular section whose corners are rounded by circular arcs of prescribed radius. The critical angle is determined by the property that a (wetting) solution surface of the capillary equation simply covering the base exists if and only if the contact angle is larger than a critical value. We were able to apply our methods developed earlier and characterize all subsidiary extremals, to give an exact result for the needed estimate.

For item (b) above, one of the two methods proposed for contact angle measurement is based ultimately on a discontinuous behavior of capillary surfaces in wedge domains. In adapting our earlier ideas to the needs of experiment design, we have been led to consider a cylindrical capillary tube whose section is a "near rhombus". An initial estimate for contact angle is to be made under terrestrial conditions, and the sectional angles adjusted accordingly. If the estimate is correct, the fluid height of any capillary surface in the tube will fall to zero (in zero g) over a region of the base, and the fluid will move to the more acute corner. If too high, the fluid will fill out both corners, and if too low the surface height will remain between explicitly known positive bounds. The method does not require precise observation of the surface in the corners. It is anticipated that very good accuracy will be obtained for contact angles greater than about 40 degrees for the wetting case.

Our expectation is that the above method will yield accuracy that is significantly better than current procedures, for values of the contact angle as above. For smaller contact angles another procedure is being studied for accurate measurement that is based on the special properties of a "two-circle" geometry. We are carrying out numerical calculations to solve the capillary equation in this geometry, and for certain values of the parameters the calculations have indicated the desired nearly-discontinuous behavior.

Further geometries have now been characterized in a study initiated in collaboration with a student, B. Fischer. Initially, the two circle configuration was modified by introduction of linear boundary segments joining the circular arcs, in order to achieve a greater thickness (and hence visibility) of the singular fluid rise. It was discovered that a very small change of that sort led to marked improvement in the predicted behavior of the surface near the critical configuration. In seeking to optimize the behavior, we are being led to investigating the possibility of a contour that admits an entire continuum of "subsidiary minimizers" for prescribed contact angle. When applied to the measurement techniques, the result could yield a more precise measurement than the previously considered procedures, for small contact angle and with less need for difficult computer calculations. In this respect the proposed method complements nicely the above described "near rhombus" procedure, which is best suited to measurement of larger angles.

Publications

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Convective and Morphological Stability during Directional Solidification

National Institute of Standards and Technology

Dr. S. R. Coriell

Dr. G. B. McFadden

Dr. B. T. Murray

Dr. J. R. Manning

Continuing Task

Objectives: The general aim of this task is the study of the fluid flow, solute segregation, and interface morphology which occur during directional solidification, including effects of gravity and microgravity. Control of solute segregation during solidification will allow preparation of materials with optimum properties. Space flight experiments, designed to determine cellular wavelengths as a function of growth conditions, are planned in collaboration with J. J. Favier, A. Rouzaud, and D. Camel of the Centre d'Etudes Nucleaires de Grenoble utilizing the directional solidification furnace being developed by the MEPHISTO project. In collaboration with K. Leonartz of ACCESS e. V., calculations of convective and morphological instability are being carried out for the succinonitrile-acetone system in support of space flight experiments on the IML-2 mission.

Research Task Description: The main focuses of this task are the interaction of fluid flow in the melt with the crystal-melt interface, and solute segregation which is controlled by interface morphology and fluid flow. In the absence of fluid flow, the conditions for morphological instability during directional solidification of an alloy at constant velocity are well established. However, for processing conditions for which instability occurs, the resulting non-planar interface morphologies (cellular or dendritic) are active areas of current research. Although progress has been made in calculating cellular morphologies, the question of wavelength selection is unresolved, and definitive experiments (without convection) are needed to provide guidance and a benchmark for theoretical developments. Numerical algorithms to calculate cellular morphologies in the absence of convection are being developed. Linear stability analyses of the effect of various types of fluid flow on the onset of morphological instability are carried out in order to delineate the role of convection. Fluid flow in the melt is calculated by a time-dependent, two-dimensional finite difference algorithm as a function of the gravitational acceleration. This allows prediction of the differences between experimental results obtained on earth and in space. The effect of time-dependent gravitational accelerations (g-jitter) are studied by linear stability calculations, asymptotic analysis, and numerical solution of the nonlinear fluid flow equations. Since the MEPHISTO furnace uses electrical pulsing and Seebeck measurements, research on the effect of electrical currents on the solidification process and solute redistribution is also underway.

Progress to Date: During directional solidification of a binary alloy at constant velocity, thermosolutal convection may occur due to the temperature and solute gradients associated with the solidification process. For vertical growth in an ideal furnace (lacking horizontal gradients) a quiescent state is possible. For a range of processing conditions, the thermal Rayleigh number is sufficiently small that the stabilizing role of the thermal field during growth vertically upwards may be neglected, and only solutal convection need be considered. In collaboration with A. A. Wheeler of the University of Bristol, the effect of a time-periodic vertical gravitational acceleration (or equivalently vibration) on the onset of solutal convection has been analyzed in the limit of large modulation frequency. When the unmodulated state is unstable, the modulation amplitude required to stabilize the system can be determined by the method of averaging, and is proportional to the modulation frequency. Comparison of the results from the averaged equations with numerical solutions of the full linear stability equations (based on Floquet theory) show that the difference is proportional to the square root of the modulation frequency. When the unmodulated state is stable, resonant modes of instability occur at large modulation amplitude. These have been analyzed using matched asymptotic expansions to elucidate the boundary layer structure for both Rayleigh-Benard and directional solidification configurations. The leading order term for the

modulation amplitude is proportional to the square of the modulation frequency with corrections proportional to the $3/2$ power of the modulation frequency. Based on these results, a detailed examination of the dependence of the stability criterion on the unmodulated Rayleigh number, Schmidt number, and segregation coefficient has been carried out.

The onset of convective and morphological instability during the solidification vertically upwards of the succinonitrile-acetone system has been calculated for a range of processing conditions and gravitational levels. For a small range of processing conditions, an oscillatory (in time) critical mode of instability is found. In addition to the numerical methods previously used, we have developed a second independent solution technique based on Chebyshev pseudospectral discretization of the governing differential equation which results in a generalized matrix eigenvalue problem for the temporal growth rate of the instabilities. This algorithm is particularly advantageous for determining oscillatory modes of instability.

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Modeling of Droplet Coalescence and Phase Segregation

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Dr. Robert H. Davis

NAG3-993 (NASA Contact: R. Balasubramaniam, LeRC)

Objectives: The primary objective of this research is to develop macrophysical and microphysical models of droplet coalescence. The objective of the macrophysical studies is to predict drop-size-distribution evolutions due to droplet collisions and coalescence during processing within the miscibility gap of bimetallic liquid-phase-miscibility-gap materials. The individual and collective action of gravitational and nongravitational mechanisms on the relative motion and coalescence of drops are considered. The objective of the microphysical research is to predict deformation, film drainage, and collision rates during the interaction of a drop with a surface or another drop due to gravitational, Brownian, or thermocapillary motion.

Research Task Description: When bimetallic liquid-phase-miscibility-gap materials, which are thought to have a variety of desirable properties, are cooled through the miscibility gap, droplets rich in one of the metals form in the liquid matrix rich in the other metal. Droplet coalescence and phase segregation then occur due to buoyancy and to thermocapillary and other nongravitational mechanisms. In order to gain a predictive understanding of these phenomena, population dynamics models are used to follow drop-size distribution evolutions in time as the droplets grow due to collisions and coalescence. Continuous drop size distributions are discretized into a large number of categories. Drops of a given mass are destroyed by coalescing with other drops and are formed by the coalescence of smaller drops. The population dynamics model tracks the formation and destruction of drops in each size category.

The relative motion of drops which gives rise to their collision and coalescence is considered to occur by gravity sedimentation, Marangoni migration, Brownian motion, and bulk flow. These collision mechanisms are considered either individually or collectively in the population dynamics models. In general, different mechanisms dominate for different processing conditions, materials properties, and drop-size ranges.

The collision kernels appearing in the population dynamics models require expressions for the collision rate between drops of two different sizes. Classical expressions attributed to Smoluchowski are improved to include attractive, repulsive, and hydrodynamic interactions between drops. In particular, trajectory calculations are used to predict collision efficiencies, which represent the ratio of the collision rate with these interactions to the Smoluchowski collision rate without these interactions, as functions of the size ratio, viscosity ratio, and other relevant dimensionless parameters. Additional studies focus on how drop deformation and van der Waals forces affect film drainage and coalescence rates.

Progress to Date:

1. A computer program has been completed for solving the population dynamics model to follow droplet size evolutions with time in homogeneous dispersions due to collisions arising from gravity sedimentation, Marangoni migration, and/or Brownian motion. Some of the key results are that a bimodal initial distribution will exhibit much more rapid coalescence due to gravity sedimentation or Marangoni migration than will a unimodal initial distribution, a unimodal initial distribution will evolve into a bimodal distribution and then into a shifted and broadened unimodal distribution, a self-similar distribution forms for Brownian coalescence, the rate of Brownian coalescence decreases with time as drops grow larger, whereas the rates of gravity-induced and thermocapillary-induced coalescence increase with time, and coalescence may be greatly reduced by antiparallel alignment of the gravity vector and the temperature gradient.

2. Collision efficiencies for Brownian motion, Marangoni migration, and gravity sedimentation for drops having a range of viscosity, thermal conductivity, and radius ratios have been computed both in the presence and absence of attractive forces. A key result is that, in contrast to rigid particles, spherical liquid drops have nonzero collision efficiencies in the absence of attractive forces. For sufficiently high thermal conductivity ratios, however, Marangoni coalescence may be prevented due to the increased velocity of the smaller drop in the temperature field surrounding the larger drop.
3. Theoretical work is in progress on spatial variations in drop-size distributions, and macroscopic phase segregation.
4. In fundamental theoretical work, the natural evolution of the approach and deformation of two drops driven together by gravity and/or van der Waals forces has been analyzed by regular and singular asymptotic expansions. In the absence of attractive forces, the drop deformation will prevent coalescence. In the presence of attractive forces, coalescence can occur due to either nose rupture or rim rupture of the film separating two drops.
5. In related experimental work, drop size distributions in transparent immiscibles are being followed with time by holography as coalescence occurs due to gravitational motion. Further experiments are underway to follow the trajectories of two interacting drops of different sizes, using video microscopy.

Publications

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Two Phase Gas Liquid Flow Under Reduced Gravity Conditions

University of Houston
Professor A. E. Dukler
NAG 3-510

Objectives: The program looks to develop an understanding of the mechanics of pressure driven simultaneous flow of a gas - liquid mixture in conduits under conditions of reduced gravity. This understanding is to be expressed in terms of a series of physical models which can be used to predict the flow characteristics such as flow pattern, pressure drop, pressure oscillations and, where applicable, bubble sizes, slug lengths and void fractions. The plan includes obtaining data from Learjet trajectories to provide insights into mechanisms and to evaluate the physical models derived. Sufficient data is to be obtained to permit the evaluation of the effect of physical properties, conduit diameter and length as well as gravity level on the behavior of the system

Research Task Description: (Experimental): Two test section sizes are being investigated, 1.27 cm dia x 114 cm long and 2.5 cm dia and 200 cm long. These are mounted in a test loop aboard the NASA Lewis Learjet which meters the gas and liquid streams, the pressure and temperatures along with the pressure gradient, the local film thickness and local cross-sectional average void fraction. Data on these time varying quantities are collected at 250 Hz and a 400 frames/sec camera is used in each trajectory to record the appearance of the flow. The time varying signals along with the visual record is used to analyze the detailed characteristics of the flow such as velocities and lengths of bubbles and slugs, flow patterns which exist and wave characteristics during annular flow.

Modelling: Separate models are to be developed for the flow pattern transitions, pressure drop in each of the patterns, bubble sizes during bubble flow and slug and bubble sizes and velocities during slug flow.

Progress to Date: (Experimental): Experimental data have been collected during 160 reduced gravity trajectories carried out on the Learjet. All of these runs utilized the 1.27 cm diameter test section. Air was used as the gas but the liquid properties included water, a water-glycerol mixture and an water surfactant mixture. Two gravity levels were explored including ± 0.02 g and 0.17 g, the latter to simulate moon gravity levels. During this period the new measuring techniques were introduced for film thickness and void fraction:

Modelling: Models have been developed for predicting flow pattern transitions at reduced gravity. A mathematical model has been developed which predicts the detailed features of slug flow including bubble and slug length as well as its propagation velocity, pressure drop and voids. This model is now under detailed test against recent data. A general model for predicting the pressure drop during bubbly flow has recently been completed and showed excellent agreement with data.

Publications

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Presentations

**Dukler, A.E., "Two Phase Gas-Liquid Flow at Reduced Gravity Conditions: Problems and Possibilities,"
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A Theoretical Study of the Influence of the Earth's Gravity and of Zero-Gravity Environment on Experimental Measurements of the Critical Properties of Fluids

University of Maryland
Professor Richard A. Ferrell

Objectives: The objective of this project is to evaluate theoretically the influence of gravity on two categories of experiments that are designed to measure the critical transport properties of fluids. In the first category is the zero microgravity light scattering experiment, scheduled relatively soon for the space shuttle. We have been working out in full detail the mechanism of fast adiabatic equilibrium, as well as the possibility of measuring the critical exponent η by light scattering. By decreasing the time required to reach thermal equilibrium in the fluid sample it will be possible to modify the protocol for the experiment so as to increase the number of data points. In the second category are some potential space shuttle experiments that are still in the planning stage, such as measurements of the critical viscosity. We intend to investigate the possibility that the goals of some of the experiments in this second category might be realizable on earth, without resort to a microgravity environment.

Progress to Date: Progress has been made on the theoretical understanding of various properties of fluids, as indicated in the following list of publications, A.1. through A.6. Of special relevance to the Zeno experiment is reference A.5., the abstract of which states,

Of fundamental importance in the fluctuation theory of second order phase transitions is the deviation of the correlation function from the mean field Ornstein-Zernike approximation, as measured by the critical exponent $\bar{\eta}$. In this note it is demonstrated how measurements of the temperature dependence of the turbidity of a fluid very close to its critical point can lead to a determination of $\bar{\eta}$. Applied to a pure fluid, this technique will require a microgravity environment.

Other results, not yet written up for publication, are the following:

1. Critical divergence of the viscosity of a pure fluid near the gas-liquid critical point. With student H. Hao we have found that the previous computations of the critical exponent are in error. When corrected, the theoretical value is in good agreement with the measured value.
2. Effect of g-jitter on the Rayleigh line width near the gas liquid critical point. Using data from shuttle flights we find this effect to be negligible.
3. Measurement of the critical viscosity of a pure fluid very near the gas-liquid critical point. By using a thin horizontal layer of fluid and allowing theoretically for the confining geometry, it may be possible to carry out ground based measurements in spite of the earth's gravity.

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Molecular Dynamics of Fluid-Solid Systems

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Dr. Joel Koplik

Dr. Jayanth R. Banavar, Pennsylvania State University

Objectives: Several interesting unresolved problems in fluid mechanics occur when the process is controlled by a microscopic region which is too small to be described faithfully by the continuum Navier-Stokes equations, or alternatively when the appropriate boundary conditions are unknown. Examples include the moving contact line singularity arising when a viscous fluid displaces a second immiscible fluid along a solid substrate, the late stages in the coalescence or fission of liquid drops, the detailed local dynamics of the spreading of liquids across a solid surface, and the freezing and structure of liquids in microporous systems. We propose to use molecular dynamics numerical simulations to study these processes. In so doing, the correct molecular behavior is built into the calculation, and the result is a relatively unbiased numerical experiment which will elucidate the above questions.

Research Task Descriptions: Beginning with standard molecular dynamics computer codes, the first new ingredient required is a molecular solid bounding wall with realistic thermal and structural properties. With this in hand, dynamics in microporous systems can be investigated by controlling the operating conditions via solid bounding walls, mimicking laboratory experiments, but with vastly higher resolution. The same type of wall can be used to examine drop spreading on substrates, and to construct a computer "four-roller mill", which will provide controlled external flow conditions for drop dynamics studies. The numerical simulations give immediately the instantaneous configurations and velocities of the fluid(s), while density and temperature profiles, average Eulerian velocity and stress fields and other continuum quantities can be obtained by averaging.

Progress to date: This research program involves several related projects, and we review the status of each in turn.

1) Vectorization and Optimization of the Numerical Code

The MD code was fully vectorized to take advantage of the vector processor facilities of the IBM mainframe at Penn State and the Cray YMP at the Pittsburgh Supercomputer Center. The program is now readily transportable and runs on all machines that we have access to. In addition to the vectorization, we implemented the layered-linked-cell algorithm of Grest, Kremer and Dunweg to speed up the program considerably. We have also written a variety of different analysis subroutines for the calculation of various quantities of interest during a run. The program has the versatility to enable any or all of these routines to be called as needed. Care has been given to allow the user to add other routines readily as the need arises.

A sub-project is underway to develop an efficient parallel Fortran code to exploit the architecture of a Connection Machine. We hope thereby to increase the computational efficiency and allow the simulation of much larger systems. At present we are experimenting with two parallel algorithms, which (for N molecules) can achieve computation times $O(1)$ and $O(\log N)$ respectively, essentially by evaluating the force on each molecule in parallel. The viability of this approach seems to depend critically on the speed of current CM hardware. Once a stable code is available, the remaining CM allocation will be applied to very large scale simulations of the problems below.

2) Development of Realistic Walls

Realistic molecular walls made of Lennard-Jones atoms have been constructed and tested. The wall density and the fluid density can be chosen at will, thus allowing the study of an incommensurate

wall-fluid system (in our older studies, this was not the case). The solid-solid potential is taken to be stronger and deeper than the liquid-liquid or solid-liquid interactions, so that the walls are nearly rigid and able to maintain thermodynamic equilibrium with the liquid. The outer layers of the walls (those away from the solid-liquid interface) are continually equilibrated at the desired temperature by a rescaling of the velocities. Detailed tests were carried out to ensure the stability of the system and the ability of the wall to act as a realistic and efficient conductor of heat. For example, use of the previous walls (made of very heavy molecules) resulted in steady heating of the fluid during a flow run, ultimately leading to the collapse of the walls. The present program instead gives a steady state situation, where the heat produced during the flow is conducted away to the walls and removed at the outer wall sink. Furthermore, the average gap-wise profile in temperature is of the expected quartic form, except for prominent superposed oscillations, which we believe is a real and new effect due to the layering of the fluid close to the walls. Similar long runs have been carried out in the case of Couette flow with shear rates more than ten times higher than what we had used previously.

3) Flow of a Single Fluid Between Walls

We have begun to study flow characteristics in the size regime where the continuum equations of fluid dynamics begin to break down. When the spacing between walls is of the order of 35 Å, the velocity profile predicted by the continuum Stokes equations is seen, but there is clear evidence of layering even in the presence of flows, as seen in the density and temperature profiles. To study the nature of the layer next to the solid surface, we have determined the two dimensional in-plane distribution function, the residence time distribution, and the structure factor. Despite of the incommensurate nature of the fluid-wall density, there is clear evidence for in-plane ordering in the first layer. Studies are underway to shrink the distance between the walls to probe the effect of confinement on the viscosity of the fluid. We are motivated here by experimental reports of a dramatic increase in the viscosity, along with the onset of non-Newtonian flow (such as shear ordering), in microporous systems which can accommodate only a few layers of the fluid. We are uniquely set up to carry out a realistic and detailed study of this phenomenon, and expect to finish this work over the summer.

4) Freezing in Confined Geometries

There is an extensive literature on the phenomenon of nucleation and freezing in bulk geometries. Molecular dynamics studies have confirmed that the classical theory which involves the formation of a critical nucleus. It has also been suggested that the presence of a "pseudo-spinodal" may cause the critical nucleus to be ramified (rather than compact) leading to some predictable departures from the conventional theory. The situation regarding freezing in confined geometries is more murky. There exist now several experiments involving light scattering, acoustic, specific heat, and neutron probes (the latter two are being carried out by colleagues at Penn State) on freezing in porous media. However, practically no theoretical work has been carried out on this subject. We have carried out some initial studies of freezing of liquids contained between walls separated by 35 Å. The results are striking and indicate a new heterogeneous mechanism of freezing. Our results suggest a two step freezing process - the liquid initially forms very distinct layers parallel to the walls, and the ordering within any layer proceeds subsequently. The layering is found to grow inwards from the wall. We have used three kinds of quench processes to study this phenomenon - an abrupt quench (in which the kinetic energies of the fluid molecules are instantaneously lowered by rescaling), a gradual quench (similar to the procedure used in bulk freezing studies), and an external quench (in which the outer wall temperature is reduced and removes heat from the liquid by thermal diffusion). Our results are in accord with recent experiments, to the extent that a comparison is possible. In the presence of rough walls, we observe that the layering tries to follow the local wall orientation, leading to stacking faults and defects as solidifying material formed near different regions of wall meet

5) Interfacial Snap-off and Coalescence

The deformation and burst of drops of liquid in shear and extensional flows have been studied theoretically and experimentally for some time, and much of the continuum aspects are understood. The initial stages of deformation can be followed quantitatively using linear stability analyses and small deformation approximations based on power series, while larger deformations can be treated using boundary-integral numerical techniques. As long as the thickness of the drop exceeds the several-thousand Angstrom range, the problem is purely one of continuum fluid mechanics. As the deformation proceeds, drops are found to take on a dumbbell shape with a narrowing neck. For highly elongated drops, allowance for the microscopic size of the neck can be made by using disjoining pressure corrections. However, when the neck thickness reaches the hundred-Angstrom range, there are too few molecules to provide a continuum, and current modeling is inadequate.

Likewise, when two drops approach in an external flow, it is known that they begin to distort, so that the adjacent spherical noses flatten out. On further approach a dimple forms, because the viscous fluid between the drops cannot escape fast enough. The two drops form axisymmetric kidney shapes, and are closest to each other along two facing circular rings. Eventually, fluctuations cause the rings to approach and burst at one point, and the fluid rapidly rearranges itself into a drop. As in the fission case, most of the process can be treated by continuum modeling, but the burst and rearrangement occurs on too short time and distance scales.

We first studied two simpler cases. The simplest interfacial instability problem concerns the capillary rupture of a cylindrical thread of liquid in equilibrium with its vapor. An ancient calculation by Rayleigh indicates an instability to long-wavelength perturbations in diameter, leading to a breakup into liquid spheres. We have observed this instability in simulated liquid threads of various sizes, in qualitative accord with theory - we show the evolution of a 3015-atom system from a cylindrical thread in equilibrium with vapor, through its rupture due to the Rayleigh instability, to its final state as a spherical drop. The dots are the positions of atomic centers, and some edges of the computational region are shown. The time scale is the natural MD time, $O(10^{-12}$ sec). Other runs with various sizes are in qualitative accord with linear theory. Although the geometry is easy to quantify, the computed time-dependent velocity and stress fields suffer from excessive thermal noise, due to the short time scales and limited system sizes. We are attempting instead to quantify the statistics of the atomic motion and displacement. A second easy calculation involves detaching a "pendant" drop of liquid from a solid support by a gravitational field. A typical case is where the drop is initially attached to a solid pin. This configuration has the advantages of an externally controlled rate (i.e., gravity) and in addition has been the subject of more precise laboratory experiments, which will assist in making comparisons. Indeed, the asymmetric shape we observe closely resembles laboratory studies of much larger systems.

For further calculations of drop deformation, we use either translation of our solid walls described above to impose a uniform shear flow, or else a "four-roller mill" to set up a better-controlled extensional flow. The former method easily develops high velocity gradients, but has the disadvantage that uniform shear entails rotation. The latter device is widely used in laboratory work, and is readily simulated using our solid algorithms above, but is not ideally compatible with the periodic boundary conditions preferred for MD calculations. This defect can be remedied at the cost of larger scale simulations, which are now in progress. We show an example of drop distortion using shear flow; the dots are the centers of the drop atoms, the drop is suspended in a background fluid which is however not displayed, and there are solid walls above and below which translate in opposite directions. In Fig. 6 we have an example of the coalescence of two drops in a four-roller mill; again the background fluid and rollers are not shown. Note that such small drops do not display the dimpling preceding rupture observed for larger drops.

6) Drop Spreading on Solid Surfaces

Recent experiments and theoretical developments, especially by the College de France group, have indicated a variety of interesting regimes are possible here, ranging from partial wetting with fixed contact angle, to precursor film advance, to terraced spreading where individual molecular layers advance. We have been simulating these phenomena by combining the molecular solids used in freezing with drops plus vapor used in fission. An example of partial wetting is where a 4000 atom drop is attracted to, and settles on, and ultimately spreads across a solid lattice. In this case the attraction between solid and liquid atoms was the same. In contrast, the solid-liquid interaction is 1.4 times more attractive, leading to the terraced wetting shown. The extreme sensitivity of the wetting to the interaction strength is remarkable. The surface tensions of the various interfaces and the resulting spreading parameter are calculated in independent simulations using the same interactions. We have found that the dynamics of the process, the evolution of drop height, radius and contact angle with time, do not appear to agree with the hydrodynamic predictions. Again, we are attempting to quantify the spreading process in terms of the atomic motion.

7) Nucleation and Bubble Growth in Solidification

Recent experiments in directional solidification by the Cummins group at CCNY have found that when typically-used materials solidify, one observes a cloud of gas bubbles formed from dissolved impurities. The bubbles form a cloud which seems to maintain a fixed separation from the advancing solidification interface. This process is of some practical interest, since such impurities can have a negative effect on the allegedly purified solid which is the commercial application of this process, and from a research point of view the bubbles can interfere with observation and measurement. We have begun MD simulations of this process, which provides a splendid combination of problems: identification and quantitative analysis of vacant regions in a dense liquid, simulation of a moving solidification interface in an imposed temperature gradient, nucleation of bubbles at an interface (heterogeneous nucleation is itself a subject with numerous open questions), and the dynamics of a bubble cloud in fluid. Initial work has developed identification algorithms, and study of the solidification process has started.

8) Molecular Dynamics Simulation of Two-fluid Phase Separation

We study the phase separation of two immiscible fluids, quenched to an unstable state within the coexistence curve in the phase diagram. This "spinodal decomposition" process has been studied recently by many experimentalists using various two-fluid systems in porous glasses. As a first step toward studying this problem using MD techniques, we work on bulk systems in three dimensions. We consider two identical Lennard-Jones 6-12 fluids, where the interaction between molecules of different species is a pure repulsive potential. A series of short-period test runs show that the system has a critical temperature at least higher than 8.0 in reduced units. We set the working temperature at 1.4, which is slightly higher than the single fluid critical temperature, and the density is 0.8 in reduced unit for each fluid.

Three 50-50 mixtures in periodic cubes of linear dimensions 18.81, 23.94 and 34.20 molecular size have been studied so far. All systems started from completely mixed states, and the separation process was studied by monitoring snap-shot pictures, single fluid radial distribution functions and their Fourier transforms. The snapshots showed that the fluids form highly connected, mutually interpenetrating networks at an early stage. As time went on, the number of branches in each network decreased. Isolated "droplets" did not play major roles in the growth of the systems we have studied. The radial distribution function initially was that of a uniform fluid, with decaying oscillations following the first major peak, reflecting the molecular discreteness. As the network grew, longer wavelength oscillations appeared at large r due to cluster discreteness. Information about the 'thickness' of the 'tubes' in each cluster network can be drawn from the location of these oscillations. On scaling the radial distance with respect to one of these positions, excellent scaling exists for $g(r,t)$ over the region of the

scale of a cluster size. The Fourier transform of $g(r,t)$, which has been checked in most previous simulation on discrete models and is the quantity analyzed in phenomenological studies of growth in spinodal decomposition, also exhibits good scaling when the contribution from cluster size scale dominates that of the molecular scale.

Growth exponents has been calculated for all three systems and the effect of different initial conditions have been checked for the smallest size and middle size systems. A tentative summary is that the growth of cluster in sizes from a few to less than twenty times of the molecule size, the exponent is between one-half and two-third. There is some evidence that the exponent is increasing as the system size becomes larger, indicating that hydrodynamic effects are more important as time goes on and the cluster size increases, as predicted by Siggia et. al. and Furukawa et. al. Further analysis is underway. This phenomenon is ideally suited for a microgravity environment - gravity is expected to play a key role in the later stages of growth. To study such effects, we have initiated gravity driven phase separation on a system of two fluids with different masses, but otherwise identical, subject to gravity forces.

Studies are also underway of the flow and freezing of diatomic molecules. These are being constructed by rigidly attaching pairs of Lennard-Jones molecules as in a dumbbell. The interplay between the rotational and translational degrees of freedom lead to rich behavior and a comparison to experiments with diatomic molecules can also be made.

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Suppression of Marangoni Convection in Float Zones by a Gas Jet

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Dr. Samuel Ohring
Work Order No. C-32007-M

Objectives: According to the amendment of September 10, 1990 to project C-32007-M, the project objectives are: Computational study of high Marangoni flows in a silicon float zone and determination of the critical Marangoni number for the oscillatory convection to occur.

Research Task Description: The onset of Marangoni convection in the float zone of liquid silicon shall be solved numerically with the aid of the Navier-Stokes equations for an axisymmetric flow with nonlinear free-surface conditions. On this free surface either the temperature gradient will be generated by heat transfer and radiation from a heater.

Progress to Date: The project was finished on July 17, 1991 as scheduled. The following summary is given on the progress in FY91 and the final results: A Navier-Stokes computer code for a general time-dependent axisymmetric float-zone model with a deformable free-surface has been developed. This code makes it possible to asymmetrically disturb the flow by moving the heater for a short time from its symmetric position between the solid surface and back. Two cases for the Marangoni numbers (based on the temperature difference between heater and melting point of silicon) $Ma = 10,400$ and $50,050$ were computed. The results show that for $Ma = 10,400$ the axisymmetric Marangoni flow is steady and stable and consists of two main rolls. For the higher Marangoni number, however, the disturbed flow becomes unstable and a persistent oscillatory mode of 0.27 Hz develops. The free surface itself is so little deformed that the assumption of a float surface is justified with no effect on triggering instability. Instability and maintenance of the oscillation is explained by the process of vortex shedding, that is, the formation of extremal vorticity in the core region of the two rolls. These two rolls interact rhythmically by alternating build-up and decay.

Publications

Lugt, H.J. and Ohring, S., "Marangoni Convection in a Gravity-free Silicon Float Zone," J. Fluid Mech. (submitted).

Stability and Instability of Thermocapillary Convection in Models of the Float-Zone Crystal-Growth Process

Georgia Institute of Technology
Professor G. Paul Neitzel
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Professor Hans D. Mittelman, Arizona State University
NAG3-568 (NASA Contact: Dr. R. Balasubramaniam, LeRC)

Objectives: The objectives of this research are an investigation of the stability properties of thermocapillary convection in a model of the float-zone crystal-growth process. Energy-stability limits provide sufficient conditions for stability of a basic state, while linear-stability limits provide sufficient conditions for instability. The successful computation of such limits for an actual float-zone basic state would identify conditions under which oscillatory convection (and hence, undesirable striations in the final material) can be avoided.

Research Task Description: The research associated with this project is being carried out in a variety of related, complementary areas: *i)* extension of energy-stability results computed for thermocapillary convection in a half-zone assuming axisymmetric disturbances to include three-dimensional disturbances; *ii)* computation of linear-stability properties of the same basic state; *iii)* application of energy-stability theory to a half-zone basic state with a deformable free surface; *iv)* consideration of alternate heat-transfer conditions at the free surface; and *v)* direct numerical simulation of oscillatory thermocapillary convection in a half zone.

Progress to Date: Linear-stability theory has been applied to the half-zone basic state previously analysed using energy-stability theory. This basic state assumes a non-deformable free surface. The assumption of normal modes renders the azimuthal dependence parametric. Discretization of the resulting partial-differential eigenvalue problem results in a generalized algebraic eigenvalue problem which is complex, non-hermitian, in addition to being indefinite as was the case with the energy theory. Several techniques for computing the eigenvalue of interest, including the direct calculation of *all* eigenvalues have been investigated. As before, a modified version of inverse iteration appears to perform best, although convergence with mesh refinement is very slow. Results computed thus far are linear-theory Marangoni numbers (sufficient conditions for instability) which lie *above* the experimental results of Velten, Schwabe & Scharmann.

Work on the application of energy-stability theory allowing for free-surface deformation is proceeding as is work on the incorporation of radiation heat-transfer boundary conditions into energy-theory analyses. At the present time, no direct numerical simulations have been performed, but we anticipate attempting some using the spectral-element code, Nekton.

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Mittelman, H.D., "Linear Stability of Axisymmetric Thermocapillary Convection in Crystal Growth," presented at the Conference on Bifurcation and Symmetry, Marburg, Germany, June 2-7, 1991.

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Dielectric and Electrohydrodynamic Properties of Suspensions

Princeton University
Dr. Dudley A. Saville
NAG8-878 (NASA Contact: R.S. Snyder, MSFC)

Objectives: This investigation focuses on understanding those electrokinetic properties of particulate suspensions related to the so-called "electrohydrodynamic effect," specifically, the dielectric constant and electrical conductivity. These properties are of crucial importance in defining the behavior of samples in various electrokinetic separation processes, especially those involving the purification of cell types.

Research Task Description: In the proposed study, experimental and theoretical work will be carried out to understand the electrokinetic properties of particulate suspensions that produce the EHD effect. Specifically, we will:

- 1) develop a body of data on the conductivity and dielectric behavior of various particulate suspensions.
- 2) develop a theory to describe how the dielectric constant and conductivity depend on the properties of the suspension
- 3) support the work at MSFC on the electrohydrodynamic distortion of samples in electrophoretic separation processes by making measurements on samples used in their program

Progress to Date: Efforts to date have focussed on preparing latex samples which have the desired electrokinetic properties, mainly by annealing suspensions above the glass transition temperature. We have prepared several samples and are currently measuring their electrokinetic properties.

Studies in Electrohydrodynamics

Princeton University

Dr. D. A. Saville

NAG 3-259 (NASA Contact: Dr. R. Balasubramanian. LeRC)

Objectives: The purpose of this work is to develop and test models of electrohydrodynamic processes involving fluid interfaces. Particular attention is given to the behavior of fluid globules in systems with poorly ionized solutes at high (applied) field strengths. An ad hoc model (the leaky dielectric) described by G. I. Taylor and J. R. Melcher will be expanded so as to include electrokinetic effects and put it on a firm theoretical basis.

Research Task Description: Extant theories which account for the details of physico-chemical processes associated with charged interfaces deal exclusively with low field strengths and fully ionized solutes. The model now used to describe processes at high field strengths -- the leaky dielectric -- omits consideration of electric double-layers, adsorption at interfaces, and chemical processes involved in the dissociation and recombination of solute species. Thus, even though the model depicts some features associated with bulk fluid motion faithfully, it fails to give a comprehensive picture.

The research involves several tasks: (i) construction of a mathematical model describing low field strength electrokinetics with fluid globules wherein the interface is permeable to ions and the electrolyte contains poorly ionized solutes, (ii) extension of the model described in (i) to high field strengths, (iii) adaptation of the model to oscillating fields and (iv) experiments involving steady or oscillating fields with fluid globules to test the theory. The experiments will take advantage of a micro-gravity environment to overcome problems associated with sedimentation and buoyancy driven convection arising from Joule heating.

Progress to Date: The sample cell and high voltage electronics have been used to study the behavior of a limited number of fluid pairs, with most of our results confined to castor oil and viscous silicon oils as host fluids with castor oil, silicon oils and water droplets. The agreement between theory and experiment is, thus far, excellent, but we have not yet been able to control the conductivity of the fluids using ionic surfactants so as to be able to study a wider range of properties. A paper describing these results has been submitted.

In addition to our studies of droplets, we have initiated a study of fluid cylinders. Here we find that an axial field can either stabilize or destabilize the cylinder, depending on the properties of the two fluids. For example, using an electric field we have been able to stabilize a cylinder whose length is more than 7 times as large as its radius; recall that the Plateau limit is $3.14 (\pi)$.

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Capillary Containment of Liquids in a Microgravity Environment: Shear-stabilization and Rupture

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NAG3-801 (NASA Contact: R. Vernon, LeRC)

June 1, 1987 - September 30, 1992

Objectives: The objective of this project is to study the influence of motion on the containment of liquids by surface tension. Modification of the capillary instability by shear-related forces is important to a wide variety of processing applications (materials and/or chemical) and is especially important in a low-gravity environment. We ask two contrasting questions: can motion enhance the stability of a configuration (containment) and what motions are generated when a configuration is unstable (collapse)? The answer to the first questions is "yes" and we explore the range of conditions under which stabilization can occur. The second questions is important, for example, to satellite droplet generation during breakup or collapse. Theory and experiment are closely tied in this work.

Research Task Description: In the low-gravity environment of a space-laboratory, without the stabilizing influence of the gravity of our experience, free or partially-free bodies of liquid must be stabilized by other means. Surface tension is the obvious candidate. The influence of motion on the containment by surface tension is important to a wide variety of processing applications (materials and/or chemical). It turns out that this influence can have surprising effects. In particular, we study the influence of liquid motion on the capillary instability. We ask two contrasting questions: can motion enhance the stability of a configuration (containment) and what motions are generated when a configuration is unstable (rupture)? The answer to the first question is "yes" and we explore the range of conditions under which stabilization can occur. The second question is important, for example, in determining the size of satellite droplets generated during breakup.

Progress to Date:

(i) has discovered and mapped out, in a variety of contexts, the windows in parameter space where hydrodynamic shear forces can stabilize capillary break-up in long cylindrical interfaces according to linear stability theory in a variety of contexts: rod flow (isothermal and thermocapillary-driven), tube flow, and core-annular rod flow (in preparation). In particular, the physical mechanism of stabilization has been identified.

(ii) has designed, built, and tested an apparatus capable of exploring the influence of shear on capillary instability in a parameter range where theory suggests stabilization may occur. Experiments for quantitative comparison with predictions are in progress. A pressure drop method of fine-tuning neutral buoyancy has been discovered and implemented.

(iii) has developed a simpler analog experimental system, the soap-film bridge, by means of which many of the fundamental influences of motion on stability may be observed and studied. The soap-film bridge has already illustrated details of the rupture phenomenon never before documented.

Publications

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Dijkstra, H.A., and P.H. Steen, "Thermocapillary Stabilization of the Capillary Break-up of an Annular Film of Liquid", J. Fluid Mech. **229** (1991).

Cryer, S.A. and Steen, P.H., "Collapse of the Soap-film Bridge: Quasistatic Description," J. Colloid & Interface Sci., 1991 (submitted).

Mechanics of Granular Materials

University of Colorado at Boulder,
Professor Stein Sture
Dr. Nicholas C. Costes, NASA/MSFC
Dr. David F. McTigue, Sandia National Laboratories
NASW-2110

Objectives: The objective of the investigation is to develop a quantitative understanding of the constitutive behavior including conditions for stability of an bifurcation in cohesionless granular materials in the dry, saturated, and partially saturated states.

Research Task Description: Granular materials, in particular, geologic deposits in their in-situ state are in most cases partially or completely saturated. In such states, the interactions between solid particles and interstitial liquids, which in turn are functions of external loading and drainage conditions during loading, can result in many catastrophic events from an engineering point of view which may involve the loss of life, property and natural resources (e.g. landslides, soil liquefaction caused by earthquakes or wave action, dam failures, land erosion, etc.) Accordingly, in view of these considerations, dry-state tests alone will yield only partial information on the constitutive behavior of cohesionless granular materials at very low effective confining pressures. For these reasons, the MGM experiment entails a test series on granular materials in dry, partially saturated, and fully saturated states.

Progress to Date: The effort started in June 1990 and we are making good progress in all areas, and we hope that both MSFC and SNL will become involved with the management of the project and apparatus design and development to a greater extent this year.

Publications

Macari, E.J.P., Sture, S., and Runesson, K., "Analysis of Low Effective Stress Characteristics of Granular Materials in Reduced Gravity," in *Proceedings of ASCE Geotechnical Engineering Special Publication* Volume II(27), 1991, pp. 1222-1233.

Klisinski, M., Runesson, K., and Sture, S., "Finite Element with Inner Softening Band," ASCE J. Engr. Mech. **117**, 576-588 (1991).

Peric, D., Runesson, K., and Sture, S., "Evaluation of Plastic Bifurcation for Plane Strain Versus Axisymmetry," ASCE J. Engr. Mech. **118**, (1992).

Sture, S., "Localized Deformation and Failure Analysis of Elasto-plastic Materials," in *Proceedings of First U.S. International Congress on Computational Mechanics*, Chicago, 1991, p 65.

Theoretical Studies of Residual Accelerations in a Microgravity Environment: Stochastic Formulation of Fluid Flow Phenomena

Florida State University
Dr. Jorge Vinals
Dr. Robert F. Sekerka
NAG3-1284

Objectives: Significant levels of residual accelerations have been detected during space missions in which microgravity experiments were being conducted. Direct measurements of these residual accelerations have shown that they have a wide frequency spectrum, ranging approximately from 10^{-4} Hz to 10^2 Hz. Amplitudes range from 10^{-6} g (earth) at the lowest end of the frequency spectrum and increase roughly linearly for high frequencies, reaching values of 10^{-4} g (earth) - 10^{-3} g (earth) at frequencies of around 10 Hz. The effect of such residual accelerations on experiments, compared to ideal zero gravity conditions, is largely unknown, especially in quantitative terms.

This research program aims at estimating the effect of the high frequency components of the residual accelerations (or g-jitter) on a variety of typical fluid experiments. First, a better knowledge of the influence of g-jitter on a given experiment will assist in its design in order to minimize or compensate for its influence. Second, tolerable levels of residual accelerations will be defined such that the processes to be studied will not be appreciably distorted by the environment in which the experiments take place. Third, errors in the quantities measured in the presence of residual accelerations will be estimated, including whenever possible some methodology for extrapolation to ideal zero gravity.

Research Task Description: The high-frequency, large-amplitude components of the residual accelerations are modeled as a stochastic or random process; that is, as a succession of random values of the intensity and orientation of the acceleration. Such an approach is expected to be relevant for a class of fluid phenomena that take place in a time scale much larger than typical changes in residual accelerations. In this limit, the behavior of the system is likely to depend on some statistical properties of g-jitter rather than on its details during a particular experiment. Our research is divided into two major parts, one analytic and the other numerical in character. In the first part, we formulate a hydrodynamic problem that explicitly includes a time-dependent gravitational acceleration modeled as a stochastic variable. Specific cases to be addressed include the motion of an interface separating two fluids of different density and corrections to diffusive processes resulting from random velocity fields induced by g-jitter. In the second part, we develop numerical algorithms to solve the governing set of stochastic differential equations and apply them to the systems described above.

Progress to Date: The effects of residual accelerations on the spectrum of excitations of a free fluid surface has been obtained. The inclusion of jitter leads to important corrections to the dispersion relation of surface waves. In the underdamped limit (the restoring force of capillarity dominates over viscous damping), the dispersion relation for propagating surface waves is modified. In the opposite limit, the decay rate of a surface perturbation is also modified. Under certain conditions, g-jitter can lead to an instability of a planar surface.

Drop Coalescence Studies

Vanderbilt University
Professor Taylor G. Wang
Dr. A.V. Anilkumar
Dr. C.P. Lee

Objectives: The objective of this project is to understand the detailed dynamics following the coalescence of initially stationary drops of the same liquid, separated by an immiscible host.

Research Task Description: The coalescence experiments are conducted in an immiscible acoustic levitator with degassed distilled water as the host medium. Different viscosity grades of silicon oil (DC 200 series) mixed with bromobenzene is the drop liquid. The mechanism involves levitating the first drop and then introducing the second drop. As the latter slowly seeks the same levitation position, the two drops come into contact with each other and coalesce. The coalescence is delayed until the host liquid between the drops is completely drained. The study of drops of very large diameter ratios has been conducted in a vertical channel with water-glycerin mixtures of various viscosities (1 - 4 cP) as the drop liquid and silicone oil (DC 200, 100 cs) as the immiscible host through which the drop translates very slowly before coalescing with the bulk liquid at a flat interface.

For drops of equal sizes there is no mixing following coalescence. However, when drop sizes are different, there is considerable mixing with the smaller drop penetrating the larger drop as a vortex. The penetration is basically driven by the capillary pressure difference between the drops and resisted by the viscous drag. In drops of finite sizes, the process is complicated by the capillary oscillations generated upon coalescence. The simpler case of coalescence of a small drop at a flat interface with the bulk of the same liquid has been studied in detail. The penetration depths following coalescence have been measured and correlated with theoretical predictions. We find that in the range of the investigation, the penetration depth is proportional to the $5/4$ th power of drop diameter and inversely proportional to the square root of the drop viscosity.

When the drop and host viscosities are comparable, drop penetration is prematurely terminated by the formation of a satellite drop. However, when the drop diameter ratio (large to small) is below a critical number satellite drop formation is inhibited and coalescence goes to completion. The influence of drop-host viscosity ratio and drop-host interfacial tension on this critical diameter ratio is being examined at the current time.

Publications

Anilkumar, A.V., Lee, C.P., and Wang, T.G., "Surface-Tension-Induced Mixing Following Coalescence of Initially Stationary Drops," Phys. Fluids, 1991 (in press).

Anilkumar, A.V., Lee, C.P., and Wang, T.G., "Momentumless Coalescence of Drops," AIAA Paper 92-0111 (in press); *30th Aerospace Sciences Meeting*, 1992, Reno.

Dynamics of Charged Free Drops Experiments

Vanderbilt University
Professor Taylor G. Wang
Dr. A.V. Anilkumar
Dr. K.C. Lin

Objectives: The objective is to develop apparatus for non-contact drop charge measurement, which will then be used to study the effect of charge on the dynamic behavior of free liquid samples.

Research Task Description: One of the major problems facing the charged drop experiment is the determination of charge quantity on a liquid sample. Although charge detection and measurement is one of the oldest experiments, there has been little improvement in the methods used for static measurement. The most frequently used instruments for non-contact charge detection are so-called field mill machines or generating voltmeters. Unfortunately, the resolution of this type of apparatus is on the order of 104 V/m and the measurement has to be done within a very short distance, typically less than one inch, while the drop charge experiment may require resolution as low as 20 V/m at a measuring distance of 6 inches. The primary objectives of this research are to address these technical challenges.

A new approach and a new device for detecting point type of charge have been developed. This device is based on the principle of reciprocal motion of a grounded conductor near a point charge. The current induced through such a process can then be used to determine the static field strength of the point charge, hence the quantity of the charge. Through proper design, very high signal-to-noise ratio can be achieved without sacrificing sensitivity.

Experiments were done to understand the characteristics of this new device. Test cases indicate that the results are in good agreement with the theoretical prediction. This device shows extremely good linearity with respect to the strength of the input static field. The resolution of the system is better than 10 V/m, which is about two orders of magnitude in sensitivity higher than other types of field measuring apparatus. The high sensitivity of this system makes it possible to conduct charged drop experiments under microgravity conditions.

Publications

Lin, K.C. and Wang, T.G., "Non-Contact Charge Measurement," Rev. Sci. Instrum., 1991 (submitted).

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4. BIOTECHNOLOGY

Biosynthesis of Cellulose Under Microgravity Conditions

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NAG9-397

Objective: Cellulose biosynthesis is an enzymatically controlled metabolic process involving two steps; (a) polymerization of glucose to form a β 1,4 glucan polymer chain; (b) crystallization of the glucan chains to form a crystalline microfibril. Because plants contain cellulose walls and respond to gravity and because cellulose biosynthesis involves a biologically-driven biocrystallization, we are interested to learn about the effects of microgravity on this process.

Research Task Description: Using the gram-negative bacterium, *Acetobacter xylinum* as the model cellulose-producing organism, we have investigated microbial cellulose assembly on the ground, in level flight, and during parabolic flight in NASA's KC- 135 airplane. During the past 14 months, we have conducted experiments on 6 separate flights. Two platforms were constructed for these experiments: (a) a shock-mounted, temperature controlled aluminum pallet on which were placed plastic Petri dishes containing colonies of bacteria for cellulose initiation and termination, and: (b) a compound microscope and video camera for observing the motion of cellulose-synthesizing *Acetobacter* cells during parabolic and level flight. Parabolic flight consisted of two basic gravity conditions, a 25 sec microgravity period, and a 20-25 sec hypergravity period ($1.5 \times g$) during pullup. The cellulose structure was analyzed by electron microscopy.

Progress to Date: We have found that during increasing exposure to the number of flown parabolas, the cellulose structure becomes more disorganized. This can be detected by negative staining assay of the cellulose microfibrils and their organization into ribbons. The distinguishing feature of this disruption is the separation or *splaying* of microfibrils within the ribbons. Cellulose synthesized on the ground and during level flight showed normal compact ribbons.

During the June 14 flight, we followed the biosynthesis of cellulose using video microscopy. The results of this experiment indicated that typical cellulose ribbon formation was occurring during parabola exposure. Thus, the splaying may be due to drying conditions during electron microscopy grid preparation. Nevertheless, the splaying probably reflects subtle differences in H-bonding of cellulose microfibrils. At this time, the effects of exposure to parabolic flight on the exact cause of splaying cannot be determined. Longer term exposure to constant conditions of microgravity will facilitate the biosynthesis of sufficient quantity of material for more detailed analyses.

Publications

Brown, Jr. R. M., Kudlicka, K., Cousins, S., and Nagy, R., "Microbial Cellulose Assembly is Altered During Parabolic Flight," Abstract, ASGSB Meeting, Washington, D.C., 1991.

Crystallographic Studies of Proteins

NASA Marshall Space Flight Center

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Objectives: The objectives of this research are to: (1) determine and refine the structure of human serum albumin (HSA) to 3.0 angstroms resolution; (2) determine and refine the structure of a novel fungal lysozyme from *Chalaropsis*; (3) determine and refine the high resolution structures of the oxidized and reduced forms of cytochrome c5 from *Azotobacter*; (4) evaluate the effects of microgravity on the growth of selected protein crystals. In addition this laboratory will provide data collection opportunities to graduate students and researchers where such facilities are not available.

Research Task Description: Protein crystallography is currently the most powerful method for the determination of the three-dimensional structure of proteins and other macromolecules. This method usually requires crystals which are relatively large (0.5 mm - 1.0 mm) in size and possess a reasonably high degree of internal order. Consequently, protein crystal growth has become the subject of an increasing number of fundamental studies in crystal growth, including several ongoing microgravity experiments. The knowledge of the three-dimensional structure of macromolecules is of fundamental importance to the field of molecular biology, and it is presently receiving considerable attention from the biotechnology industry based on its promising potential for application in rational drug design and protein engineering.

We have constructed a diffraction laboratory for evaluating our results from ground-based experiments and our participation in flight experiments manifested on STS-26, STS-29, STS-31, STS-32. In addition to the evaluation of the x-ray diffraction properties of protein crystals, this laboratory is utilized to determine the three-dimensional structures of several challenging problems in molecular biology.

Crystals of several important proteins have been grown in our laboratory including a new crystal form of HSA, *Chalaropsis* lysozyme, cytochrome c5, the Fab portion of the antibody expressed against gp41 of the human immunodeficiency virus (HIV-I), and Interleukin 6. HSA is the most abundant protein of the circulatory system. There it plays several major roles in the transport, distribution, and metabolism of a diverse variety of endogenous and exogenous ligands. The three-dimensional structure of HSA was determined for the first time in our laboratory. The detailed atomic structure will provide a wealth of information regarding the many remarkable properties of this protein. *Chalaropsis* lysozyme is an enzyme which displays a broad spectrum bacteriolytic function and apparently belongs to a novel lysozyme structural class. A detailed understanding of the molecular structure will explain the chemical basis of the bacteriolytic function and could provide important information for future experiments in biotechnology. The crystals of cytochrome c5 diffract x-rays to very high resolution (approximately 1.0 angstrom) and will provide further insight into the mechanisms of electron transport. The structure of the HIV Fab will reveal important information concerning antibody/antigen interaction and offers additional insight into the design of new recombinant therapeutic antibodies. Interleukin 6 (IL-6) appears to be a key protein in the modulation of the immune system. Details of the IL-6 structure will provide important

insights into the function of this extremely important cytokine. The structure determination and refinement of these and other protein crystals is actively in progress.

Progress to Date: The structure of HSA has been successfully determined and refined to a resolution of 3.2 angstroms. Crystals of several other serum albumins with enhanced quality have been successfully grown in our laboratory over the past year. Preliminary assessments of the quality of human serum albumin crystals grown in microgravity and by gel methods have revealed enhancements in the resolution. Further investigations of the differences between ground-based and flight crystals will be addressed pending additional flight opportunities. The atomic coordinates obtained from the human serum albumin crystallographic studies are now being applied to understand the detailed chemistry of serum albumin. Additionally, several other serum albumin structures have now been determined by the molecular replacement method to better than 3.0 angstrom resolution. Some of these structures are still in the refinement stage until the complete amino acid sequences have been determined. Comparisons of these structures will reveal a wealth of information regarding the evolution and chemistry of this important macromolecule. Details of these studies will be published in the near future.

The three-dimensional structure of a human monoclonal antibody (Fab 3D6), which binds specifically to the transmembrane protein gp 41 of the human immunodeficiency virus type I (HIV-1), has been determined by crystallographic methods to 2.7 angstroms. The antibody belongs to the subclass IgG1 (kappa) and exhibits antibody dependent cellular cytotoxicity. The quaternary structure of the Fab is in an extended conformation with an elbow bending angle between the constant and variable domains of 175 degrees. Hypervariable loop H3, residues 102H to 109H, is unusually extended from the surface. The complementarity determining region forms a hydrophobic pocket which is created primarily from hypervariable loops L3, H3, and H2. Studies with a small antigenic peptide suggest that this binding pocket recognizes the putative solvent exposed disulphide loop CSGKLC of gp 41. Although numerous Fab structures have now been determined by crystallographic methods, this is the first representative structure of a human monoclonal antibody and the first structure of a human antibody expressed against a major epitope of the AIDS virus. This structural work is part of a series which aims to elucidate the detailed nature of human monoclonal antibodies expressed against the AIDS virus together with their respective antigenic complexes. The initial work on the Fab 3D6 has been submitted for publication.

The high resolution structure of cytochrome c5 is currently under refinement at 1.35 angstroms. This work is now being conducted in conjunction with the NASA JOVE program.

Publications

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L. J. Delucas, C. D. Smith, H. W. Smith, S. Vijar-Kumar, S. E. Senadhi, S. E. Ealick, D. C. Carter, R. S. Snyder, P. C. Weber, R. Salemme, D. H. Ohlendorf, H. M. Einspahr, L. L. Clancy, M. A. Navia, B. M. McKeever, T. L. Nagabhushan, G. Nelson, A. McPherson, S. Koszelak, G. Taylor, D. Stammers, K. Powell, G. Darby, and C. E. Bugg "Protein Crystal Growth Results for Shuttle Flights STS-26 and STS-29," J. Cryst. Growth **110**, 302-311 (1991).

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Miller, T.Y., He, X.M., and Carter, D.C., "A Comparison Between Protein Crystals Grown with Vapor Diffusion Methods in Microgravity and Protein Crystal using a Gel Liquid-Liquid Diffusion Ground-based Method," J. Cryst. Growth, 1991 (submitted).

Protein Crystal Growth in Low Gravity

Stanford University

Robert S. Feigelson

Robert C. DeMattei

October 1990 - October 1991

Objectives: The ultimate objective of this research program is the careful design of an experiment to study the effect of low gravity on the growth of protein crystals in a long-duration space flight. The proper design of the flight hardware and experimental protocols are highly dependent on understanding the factors which influence the nucleation and growth of crystals of biological macromolecules. Thus, the primary objective of this research is centered on investigating these factors and relating them to the body of knowledge which has been built up for "small molecule" crystallization. This data also provides a basis of comparison for the results obtained from low-g experiments.

Research Task Description: This research program was designed to study the mechanisms of protein crystal growth and the parameters which influence growth and crystal perfection. Canavalin was chosen as a model protein in initial studies which included 1) a determination of its solubility diagram, 2) a study of the growth rate of canavalin crystals, 3) a determination of the growth mechanism, and 4) a study of the fluid flow behavior around growing protein crystals. These tasks have been completed and published.

The current scope of the program has been directed toward other protein materials including lysozyme and isocitrate lyase and new processing techniques designed to control nucleation and growth independently. This part of the program now includes the following subjects:

- 1) To determine the relationship between gravity and the morphology and quality of isocitrate lyase crystals. This involves determining the factors which cause isocitrate lyase to grow as dendritic crystals in 1-g and as more equi-axed crystals in μ g.
- 2) To develop a method for controlling the nucleation and growth processes independently through the use of localized supersaturation control.
- 3) To develop a predictive model for protein crystal growth.
- 4) To develop conceptual designs for long-term space flight experiments based on the data gathered by this program.
- 5) To develop an understanding of the relationship between growth rate and crystal perfection.

Progress to Date: The successful results on the growth of isocitrate lyase (ICL) in recent space experiments led us to study the reasons for this dramatic improvement in crystal morphology and quality. Using the well known hanging drop method, isocitrate lyase crystals grow with a "dendritic" morphology under 1-g conditions and as equi-axed crystals in μ g. The investigation of this behavior was undertaken as a joint project with DuPont.

The usual expectation is that crystal growth benefits from a low-g environment because of reduced solutal convection and the absence of sedimentation. Observation of ICL crystals growing in hanging drops (1-g) clearly showed that the poor morphology was not the result of sedimentation. A sitting drop cell was therefore constructed at Stanford to allow schlieren imaging to study the effect of solutal convection on crystal morphology. While convection was not observed, the crystals grew with equi-axed morphology similar to those grown in space. This result was believed to be due to the design of the cell which limited the solution surface area available for evaporation. To further study the effect

of the solution surface area on the morphology of ICL crystals, crystals were grown in capillaries of various diameters. The best ICL crystals were grown in 1.88mm dia. capillaries and showed an "octagonal" morphology. Diffraction studies (DuPont) of all three types of crystals show the same space group (orthorhombic) and only slight changes of the unit cell parameters. The changes in external morphology seem to be related to the rate of equilibration of the crystallizing solution. The models of Sabille et al. (J. Crystal Growth, 110 (1991) 72 and 80) which relate the elapsed time to the volume during crystal growth by evaporation in the hanging drop and in capillaries were applied to the growth of ICL. The results showed that the small drops (4 μ l) used in most of the 1-g experiments evaporate more rapidly than the larger drops (30 μ l) used in the space experiments and that the capillary evaporation rate was significantly slower than either of these. By using the solubility data for lysozyme (Pusey, private communication) and modifying the model to account for the effect of growing crystals in the solution, it was possible to show that, for the same initial volume of solution, the supersaturation (the driving force for nucleation and growth of crystals) reached a peak which was twice as great in the drop as in the capillary. The results of applying these models supports the conclusion that the rapid equilibration of the hanging drop in 1-g leads to high degrees of supersaturation and, thus, to unstable growth.

Fowles et al. (J. Crystal Growth 90 (1988)) have observed that the evaporation rate of hanging drops in μ g is slower than that on Earth. They attributed this to the formation of a "skin" of concentrated solution at the drop's surface. Comparing the evaporation rate of capillaries with the meniscus upwards and downwards indicated that the evaporation rate was slightly slower when the meniscus was downward. A layer of precipitate (the protein) was observed to form when the meniscus was downward which further confirms Fowles' observation. This layer also acted as a sink for the protein lowering the supersaturation and promoting better crystal morphology.

The studies with ICL highlight the importance of the effect of growth rate on crystal quality. This will be a new area of emphasis in this research program. A plan is being developed to study the effect of growth rate not only on macro-defects such as external morphology, but also on micro-defects which can reduce the x-ray diffraction resolution of the crystals. Techniques will be developed to reduce these defects.

A large scale mockup of the proposed space flight experiment has been built to test the ability to control the nucleation and growth phases of crystallization independently. The mockup consists of a temperature controlled chamber which holds the growth cell. The bottom of the cell has a temperature controlled spot (0.6 mm dia.) which is used to thermally induce nucleation. Temperature profiles taken with the spot at 10°C and the chamber at 25°C showed temperature gradients as high as 200°C/cm. This device (Thermonucleator) has been used to nucleate and grow crystals of ice, Rochelle salt and lysozyme.

The challenge in using this approach to controlling protein crystallization is to limit the number of nuclei formed. The present method is to lower the spot temperature to produce a 400% supersaturation (12°C with 37mg/ml lysozyme, pH 4.0 and 2% NaCl with a bulk temperature of 20°C) and allow the crystals to nucleate and grow. The temperature of the solution and spot are then raised and all but a few of the crystals are dissolved. The remaining crystals are then regrown at lower supersaturations. This approach has produced some large crystals (200-300 μ).

Publications

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Morphological Differentiation of Colon Carcinoma Cell Lines in Rotating Wall Vessels

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Objectives: The objectives of this project are to determine whether 1) microgravity permits unique, three-dimensional cultures of neoplastic human colon tissue and 2) this culture interaction produces novel intestinal growth and differentiation factors.

Research Task Description: The initial phase of this project will test the feasibility of microgravity for the cultivation and differentiation of human colon carcinoma. We propose to do this in rotating wall vessels (RWVs) which provide a low shear stress environment in unit gravity. In this environment, early experiments have demonstrated normal human colon fibroblasts stimulate the differentiation of certain human colon carcinoma cell lines so that they produce three-dimensional tissue masses that are similar to neoplasms in patients or in xenografts in athymic nude mice. The important question is whether this differentiation induced by fibroblasts is due to the low shear stress environment of the RWV or whether this will also occur in conventional unit gravity culture. Experiments will compare microcarrier suspension cultures in the RWVs to similar cultures in standard culture systems in unit gravity. Should the low shear stress environment of the RWV be superior to that of conventional culture systems, then the co-culture experiments should be attempted in an actual microgravity environment.

Progress To Date: This project started in June of 1991 and we received RWVs a month later. We are surveying new human colon carcinoma cell lines for growth in the RWVs to determine whether any of these lines are better than the original cell lines for study. In addition, we are initiating studies into the effects of suspension culture on cell adhesion. Induction of differentiation may alter cell recognition and attachment which is reflected in changes in cellular adhesion to basement membrane and extracellular matrix proteins. Results to date are too preliminary for comment.

Publications

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Separation of Chromosomes-size DNA Molecules

University of Pennsylvania

Professor Ponzy Lu

Dr. Young Cho, Drexel University

Dr. Lee Silver, Princeton University

NAG 8-748 (NASA Contact: R. Snyder, MSFC)

Objectives: This project addresses the problem of DNA separation by gel electrophoresis. Since we are interested in DNA molecules 10^6 - 10^8 base pairs in length (10^9 - 10^{11} daltons), or molecules that are polyanions of 2nm diameter with lengths of millimeters to many centimeters which distort as they move through the electrophoretic matrix, existing theories and models for electrophoretic transport processes must be modified.

Research Task Description: This effort will complement parallel national efforts to map and sequence the entire human genome. There are currently limitations in the human genome project which can be addressed by the technologies to be developed by this research project. Even if the human genome project is completed using existing methods, the technology to be developed here will extend the benefits of those efforts to all biological species, including those used as models of human disease and for agriculture and industry.

This project is an extension of NASA support of electrophoretic separation process development over the past decade. The extrapolation of currently employed methods for DNA separation suggests the use of more dilute electrophoretic media to allow larger porosity and consequently shorter reorientation times of the polymer subjected to pulsed electric fields. The only method to maintain fluid stability in the presence of chemical and physical density heterogeneity is the microgravity environment. Since the separated DNA will be used for genetic analysis, this program will keep space science applications at the cutting edge of biotechnology.

Progress to Date: The investigations follow that outlined in the original proposal with emphasis on identifying the microgravity component. We have modeled the motion of DNA in Newtonian and non-Newtonian fluids using thin wire as a mechanical model, with gravitation as the electrical field analog. The results were unexpected and were used to formulate the experiments with DNA. The findings show that the wire falls end first, axis vertical, in the non-Newtonian fluid and longer wire falls faster. In the Newtonian fluid all wire lengths fall at the same rate, horizontally. This latter observation parallels the observations of Olivera, Blaine and Davidson [Biopolymers 2, 245-257 (1964)] where DNA of all length move at identical velocities in free Newtonian fluids in an electric field.

The apparatus, located at the University of Pennsylvania, to directly observe the orientation of DNA molecules during the electrophoretic process was built in year 1 and has been used to observe DNA orientation in electric fields typically used for gel electrophoresis. The experiments show considerable DNA orientation in non-Newtonian compared to Newtonian fluids. In fact, the orientation data suggest that the DNA molecular configuration in un-crosslinked polyacrylamide is similar to that in conventional agarose electrophoresis. Since our non-Newtonian fluid is a non-crosslinked polyacrylamide, the need for microgravity to prevent convection becomes obvious. Given the length of naturally occurring DNA, the dimensions of the electrophoresis system would need to be measured in meters rather than centimeters making Earth bound stabilization by density gradient less useful, if not useless. An absolute requirement for convection free fluid is necessary for resolution in the separation process.

As an alternative matrix for the DNA separation process, glass bead material described in the original proposal was explored. Experiments are in progress to establish that megabase lengths of DNA are indeed electrophoresed gels in the presence of glass beads of 170-200 μm diameters.

Publications

Cho, K., Cho, Y.I., and Park, N.A., "Studies on the Hydrodynamics of a Vertically Falling Thin Cylinder, Part A: A Purely-viscous Non-Newtonian Fluid," J. Non-Newtonian Fluid Mech. (submitted).

Cho, K. and Cho, Y.I., "Studies on the Hydrodynamics of a Vertically Falling Thin Cylinder, Part A: A Viscoelastic Fluid," J. Non-Newtonian Fluid Mech. (submitted).

Cho, K., Chee, D-W., and Cho, Y.I., "A Study of the Length Effect on the Mobility of a Thin Cylinder in a Free Viscoelastic Solution," J. Non-Newtonian Fluid Mech., in preparation.

Chee, D., Cho, Y.I., and Lu, P., "Electrophoretic Orientation of Lambda DNA in Linear Polyscrylamide Solution," Nucleic Acids Res., in preparation.

Presentations

Cho, K., Cho, Y.I., and Park, N.A., "Hydrodynamics of Large-Aspect Ratio Circular Cylinders Moving Vertically in a Viscoelastic Fluid," *ASME Winter Annual Meeting*, Atlanta, December 1991.

Cho, K., Cho, Y.I., and Park, N.A., "Study of Large-Aspect Ratio Cylinders in a Viscoelastic Solution," *63rd Annual Meeting of Society of Rheology*, Rochester, NY, October 1991.

Studies into the Protein Crystal Nucleation Process

NASA Marshall Space Flight Center
Dr. Marc Lee Pusey

Objectives: The objectives of this research program are to characterize and understand the aggregation processes which occur during the nucleation and crystallization of protein crystals. Initially, these studies are being done using hen egg white lysozyme as the model protein. Studies into other proteins will be initiated when the techniques and knowledge base for lysozyme are sufficiently well developed. The scope of the studies currently under way includes; the determination of solubility diagrams as a function of a wide variety of solution parameters; the determination of the aggregation pathway, and the kinetics and equilibria for each succeeding stage of that pathway; studies into the solvent (water and any added co-solutes) interaction with the protein molecule and its effect on the aggregation process. Additional results from these studies, over and above the determination of the aggregation path-way, will be an increased understanding of both the solubilization and desolubilization of protein molecules, and the role of the added solute components in this process.

Progress to Date: The aggregation of lysozyme was originally to be studied using light scattering intensity, isothermal calorimetry, and covalent cross-linking techniques. We have subsequently determined that light scattering techniques will not give the sought after information, and that some other method will have to be used. As a result, we have developed a dialysis kinetics technique which will enable us to directly measure aggregate sizes and concentrations in both undersaturated and nucleating solutions, a feat which light scattering is totally incapable of doing. We now have direct measurements of the monomer concentration in lysozyme solutions up to 9 fold saturation. Results obtained so far have supported our initial postulate that the solutions are highly aggregated, and that crystal growth proceeds by addition of aggregates pre-formed in the bulk solution.

We have begun using fluorescence techniques to further study the protein-solvent interactions and the fate of the solvent molecules during nucleation. The results obtained show that many more Cl⁻ ions (increasing concentrations of which are used to initiate nucleation and crystal growth) are found in the immediate vicinity of the protein than there are basic groups to account for them. We tentatively assigned these molecules to the hydration shell about the protein molecule. During the aggregation steps of nucleation, we have been able to directly observe the subsequent release of these molecules as the hydration shell was removed from the contacting regions. This data, obtained by fluorescence techniques, correlated very well with light scattering intensity kinetics data for following the protein aggregation process. A microcalorimeter has been assembled to the point where it can be used in preliminary experiments. When the nucleation process is followed this way, the initial binding of Cl⁻ ion onto the protein shows up as a very large exotherm. Interestingly, we as yet are unable to optically follow this reaction using fluorescence due to the rapidity of the reactions involved. Subsequent to the protein-Cl⁻ binding reaction a low, slow exothermic reaction was observed which corresponds to the aggregation reactions observed by fluorescence and light scattering. We believe that the major exothermic process for the process protein → protein.Cl⁻ → protein.crystal occurs during the binding of Cl⁻, and that comparatively the replacement of the protein-Cl⁻ interactions with protein-protein interactions has a low ΔH . Accurate phase data are the bedrock upon which investigations of phase changes are built. Using the solubility column technique developed in this laboratory, we have now finished the tetragonal lysozyme solubility diagram from pH 4.0 to 5.4, and from 2.0 to 7.0 % NaCl. We have since initiated work on the orthorhombic solubility diagram over the same range of conditions, and expect that to be completed by the middle of 1992. Also, comparative solubility studies on the Br⁻ and I⁻ salts of lysozyme are now underway.

Publications

Cacioppo, E., Munson, S., and Pusey, M. L., "Protein Solubilities Determined by a Rapid Technique and Modification of that Technique to a Micro-method," J. Cryst. Growth **110**, 66-71 (1991).

Pusey, M. L., "Estimation of the Initial Equilibrium Constants in the Formation of Tetragonal Lysozyme Nuclei," J. Cryst. Growth **110**, 60-65 (1991).

Pusey, M. L. and Munson, S., "Micro-apparatus for Rapid Determinations of Protein Solubilities," J. Cryst. Growth, 1991, in press.

Cacioppo, E. and Pusey, M. L., "The Solubility of the Tetragonal Form of Hen Egg White Lysozyme from pH 4.0 to 5.4," J. Cryst. Growth, 1991, in press.

Pusey, M. L., "Continuing Adventures in Lysozyme Crystal Growth," J. Cryst. Growth, 1991, submitted.

Wilson, L. J. and Pusey, M. L., "Determination of Monomer Concentrations in Crystallizing Lysozyme Solutions", J. Cryst. Growth, 1991, submitted.

Sibille, L. and Pusey, M. L., "Investigation into the Early Stages of Lysozyme Crystallization," J. Cryst. Growth, 1991, submitted.

Cacioppo, E. and Pusey, M. L., "The Effects of Acid Treatment and Calcium Ions on the Solubility of Concanavalin A," J. Cryst. Growth, 1991, submitted.

Advanced Protein Crystal Growth

University of Alabama in Huntsville
Center for Microgravity and Materials Research
Dr. Franz Rosenberger
NAG8-868 (NASA Contact: Dr. R.S. Snyder, MSFC)
March 1991 - March 1992

Objectives: The inability to routinely grow crystals of sufficient size and quality for diffraction studies is considered the major bottleneck in the further widespread development of the field of protein crystallography. By comparison with inorganic crystal growth, the fundamental understanding of the dynamics and kinetics of protein crystallization is still rather rudimentary. Correspondingly, the existing protein crystal growth technology provides only for limited control of essential growth parameters such as supersaturation and, consequently, the number and timing of nucleation events, and the growth rate. Most protein growth experiments to date result in a large number of small crystals. In low gravity growth experiments these uncontrolled growth conditions can become somewhat mitigated. The reduction of buoyancy-driven, solutal convection reduces the probability of concurrent nucleation in the vicinity of initially formed crystallites. This may be one of the reasons for the increase in size and decrease in the number of protein crystals that have been obtained in space.

The objectives of this research program are (a) the determination of the advantages and drawbacks of using temperature as a control parameter in protein crystal growth; (b) the development of a semi-automated protein crystal growth technology that excludes further nucleation after select initial nucleation or seeding, and provides for the controlled, non-interfering growth of the initial nuclei or seeds.

Research Task Description:

1. Development of a semi-automated technique to determine solubilities of proteins as a function of temperature in drop-size solution volumes.
2. Application of this technique to various proteins of interest for crystallization experiments under reduced gravity conditions.
3. Development of a technique for protein crystallization in x-ray diffraction capillaries, utilizing the solubility data.

Progress to Date: Progress has been excellent. We have been able to show that the temperature dependence of protein solubilities can readily and accurately be determined with a scintillation (light scattering) technique in solution volumes as small as 100 μ l. Solubility data have been determined semi-automatically for lysozyme and horse serum albumin, with the latter showing pronounced retrograde solubility. These data have been advantageously used for the control of nucleation and growth of protein crystals through temperature variations. Specifically, we have succeeded in growing lysozyme and horse serum albumin single crystals of sufficient size inside glass capillaries used for x-ray diffraction studies, using solution volumes of about 30 μ l.

Growth of DNA Crystals in a Microgravity Environment

University of Pennsylvania

Dr. Donald Voet

K. Swaminathan

NAG8-770 (NASA Contact: K. D. Sowell, MSFC)

July, 1990 - June 1991

Objectives: The objective of the research is to determine the X-ray structure of several large segments of double stranded DNA. To this end, it is necessary to crystallize the DNA. Since DNA is difficult to crystallize in a form suitable for X-ray diffraction, we are proposing to study the effects of microgravity on DNA crystallization.

Research Task Description: We intend to synthesize and crystallize several non-self complementary segments of B-DNA in the range 12-20 base pairs. The crystals, when obtained, will be used to determine the X-ray structure of the DNAs in order to establish how the DNA conformation varies with its base sequence.

Progress to Date: We have synthesized a number of different species of non-self-complementary double-stranded DNA ranging in size from 10-20 base pairs. We have developed new methods of purifying these substances. Four of them seem particularly promising candidates for crystallographic studies although one of these crystals are yet suitable for x-ray diffraction analysis. Three of the DNAs, which consist of 12, 18, and 20 base pairs, contain segments of the *E. coli lac* operator. The fourth is a 10 bp segment containing the TATA box sequence. All of these DNAs are favorable candidates for microgravity crystallization trials.

5. GLASSES AND CERAMICS

Kinetics of Phase Transformations in Glass Forming Systems

Washington University
Professor Kenneth F. Kelton
NAG 8-873 (NASA Contact: E. Ethridge, NASA/MSFC)
April 16, 1991 - April 15, 1992

Objectives: The objectives of this research are to develop computer models for analyzing calorimetrically obtained nucleation and crystallization data, and to design and carry out experiments to establish the validity of these models. The successful completion of these studies will provide an improved understanding of nucleation and crystallization phenomenon occurring in glass forming systems, explain anomalous kinetic parameters often obtained from crystallization experiments, and allow a more quantitative analysis of calorimetric data. These new techniques will allow real-time experiments of phase stability and transformation in a micro-gravity environment.

Research Task Description: The computer modeling, some of the experimental calorimetry studies, and related microscopy studies, will be carried out at Washington University. Initially, lithium-disilicate glass is being studied, since most of the thermodynamic and kinetic parameters are known; the study will be expanded to other silicate glasses in coming years. The preparation of the silicate glasses and much of the experimental calorimetric measurements will be carried out by Dr. C. Ray and Dr. D. Day of the University of Missouri, Rolla, under a related contract.

Progress to Date: Funding for the work at Washington University commenced in May, 1991. The work is proceeding well; it is ahead of the schedule originally proposed. The computer models that describe nucleation and growth by simulating directly, the evolution of the nonequilibrium cluster distribution, have been successfully written and tested. Using fundamental kinetic and thermodynamic parameters, these models can : (a) prepare a glass by directly simulating the quench, taking into account the development of the cluster populations and their effect on the nucleation rates; (b) anneal that glass for any desired population; and (c) simulate the crystallization of these glasses under isothermal or nonisothermal conditions (duplicating the conditions in thermal analysis experiments) without making the usual restrictive assumptions. These models will now be applied (1) to test the effects of the quench rate and preannealing treatments on the crystallization of glasses, (2) to probe the effects of particle size and nucleating agents, (3) to probe the validity of existing analytical expressions used to analyze nonisothermal transformations, and (4) to design new methods for making nucleation and crystallization measurements.

We are already carrying out calculations related to points (1), (3) and (4). A paper discussing the general validity of a recently suggested method for estimating nucleation rates by measuring the heights of nonisothermal DSC scans, following preannealing treatments at different temperatures, is currently being written. A new graduate student is carrying out calculations to test the validity of generally used expressions for nonisothermal transformations

Publications

Kelton, K., "Estimates of the Nucleation Rate from Nonisothermal DSC and DTA Measurements," J. Non-cryst. Solids (in preparation).

Use of Microgravity to Improve the Efficiency and Power Output of Ni-Doped Laser Glasses

University of Missouri-Rolla

Dr. Chandra S. Ray

NAG8-779 (NASA Contact: E. Ethridge, MSFC)

July 1, 1989 - June 30, 1991

Objectives: Glasses prepared in microgravity are reported to be more chemically homogeneous and to contain fewer impurities than glasses prepared on earth. These two advantages can be used to prepare laser glasses in microgravity with a more uniform distribution of the laser active ions (Nd^{3+} in this case) and negligible amount of impurities. The resulting effects would be an increase in the limit for concentration quenching for Nd^{3+} ions and a decrease in threshold energy for laser action. The objectives of this research are to, therefore, (1) increase the concentration of Nd^{3+} ions in various glasses prepared in microgravity before concentration quenching becomes important, thereby, preparing laser glasses with increased output power per unit volume, (2) decrease the threshold energy and further increase the output power by suppressing or eliminating impurities in the glass, and (3) compare selected properties such as fluorescence efficiency (ratio of input to output energy), fluorescence lifetime, and bandwidth for the laser glasses prepared in microgravity and on earth.

The anticipated results from this study will provide quantitative data showing the extent to which the power efficiency is increased and the threshold energy is decreased for the laser glasses prepared in microgravity. The practical use would be to take advantage of the higher compositional homogeneity and chemical purity for glasses prepared in microgravity to produce laser glasses that would yield more output power per unit volume than the glasses melted on earth.

Progress to Date: Laser glass compositions in the silicate and phosphate systems, which would be suitable for flight experiments, were determined on the basis of their melting temperature, glass forming tendency, and related properties. Instrumentations required to measure the fluorescence intensity, η , and fluorescence lifetime, T_f , for the Nd^{3+} fluorescence at 1060 nm ($4F_{3/2} \rightarrow 4I_{11/2}$ of) were designed and constructed, η and T_f for the silicate and phosphate laser glasses mentioned in (1) above were determined using this apparatus. The limits for concentration quenching for the same silicate and phosphate glasses were determined from absorption measurements and using Judd-Ofelt theory. Selected other properties such as radiative lifetime, fluorescence line width, induced emission cross section, refractive index, density, and chemical durability for these glasses were also determined as a function of Nd_2O_3 concentration.

Experiments directed towards the improvement of laser properties by suppressing the impurities and improving the homogeneity for glasses processed in microgravity could not be completed due to time limitations and unavailability of flight opportunities. To investigate and accomplish these objectives, a "renewal proposal" requesting flight opportunities has been submitted to NASA.

Publications

Xu, X.J. and C.S. Ray, "The Optical Properties of Neodymium Ions in the Li-Ca-Silicate and Phosphate Glasses," in *Proceedings of 93rd Annual Meeting of the American Ceramic Society*, 1991, in press.

Glass Formation and Crystallization Behavior

University of Arizona
Michael C. Weinberg
George F. Neilson

Objectives: The objectives of this program are the analysis of the crystallization behavior of marginal glass-forming systems (with the goal of assessing the possibility of containerless experiments), the experimental investigation of the crystal nucleation and growth of simple, model glasses, and the theoretical interpretation of experiments used to determine nucleation and crystallization rates.

Research Task Description: Crystal nucleation may be enhanced by the presence of foreign substances in contact with the melt. Since crucible walls and contaminants introduced into the melt from the crucible can serve as heterogeneous nucleation sites, an uncontained melt might be subject solely to homogeneous nucleating conditions. However, in order to assess the potential advantages of containerless processing at least two items are needed: (1) comparative ground based nucleation and crystallization data, (2) a more comprehensive knowledge of the factors which influence the crystallization behavior of glasses and reliable theory to explain and interpret such ground based experiments. It is the purpose of this program to address some of these issues which will allow for judicious choices of glasses for flight experiments and enable one to interpret their results.

Progress to Date: We have continued theoretical analyses of certain aspects of the standard description of thermoanalytical crystallization experiments. In particular, an analysis was made of two schemes which have been proposed for the determination of homogeneous crystal nucleation rates in glasses via DTA measurements. One method is based on the assumption that the inverse of the temperature at which the DTA crystallization rate is maximum will always increase as the number density of nucleated particles increases. The other method is founded on the assumption that the intensity at the peak height always increases as the peak height temperature increases. Theoretical studies of heterogeneous crystal nucleation have continued, too. A simple model was used to describe the heterogeneous nucleation process which assumed that the substrate particle must attain a critical size prior to its being effective as a nucleating agent. The model was applied to discuss some recent findings in the Raman Scattering of heated lithium disilicate glass. It was shown that the anomalous scattering behavior could not be taken as evidence for the formation of lithium disilicate crystals on metastably formed lithium metasilicate crystallites.

On the experimental side, the nucleation and crystallization behavior of several marginal glass-forming compositions were investigated. The preparation and behavior of several calcium aluminate compositions, prepared by sol-gel synthesis, were reported. In addition, the crystallization behavior of these materials was studied, using bulk samples as well as thin gel films on silica slides. These compositions are of interest as a consequence of their use as mid-IR fiber material. Homogeneous crystal nucleation was investigated in lithium diborate glass. It was found that neither the magnitude nor the temperature dependence of the nucleation rate could be described by Classical Nucleation Theory.

Publications

Goktas, A.A., Neilson, G.F., and Weinberg, M.C., "Crystallization of Lithium Borate Glasses," J. Mater. Sci. (accepted).

Weinberg, M.C., "The Use of Site Saturation and Arrhenius Assumptions in the Interpretation of Non-Isothermal DTA/DSC Crystallization Experiments," Thermochim. Acta (accepted).

Smith, G.L. and Weinberg, M.C., "Crystal Nucleation in Lithium Diborate Glass," Phys. Chem. Glasses **32**, 37 (1991).

Weinberg, M.C., "On the Analysis of Non-Isothermal Thermoanalytical Crystallization Experiments," J. Non-Crystalline Solids **127**, 151 (1991).

Weinberg, M.C., "Heterogeneous Nucleation on Developing Substrate Particles," Phys. Chem. Glasses **32**, 168 (1991).

Goktas, A.A. and M.C. Weinberg, "Preparation and Crystallization of Sol-Gel Calcium Aluminate Compositions," J. Amer. Ceram. Soc. **74**, 1066 (1991).

Weinberg, M.C., "The Interpretation of DTA Experiments used for Crystal Nucleation Rate Determinations," J. Amer. Ceram. Soc. **74**, 1905 (1991).

Weinberg, M.C., "Crystal Growth and Interfacial Transport in Classical Nucleation Theory," Phys. Chem. Glasses **31**, 44 (1990).

Chen, A., Neilson, G.F., and M.C. Weinberg, "Glass-Forming Ability of a Calcium Niobate Composition by Sol-Gel and Conventional Preparation Methods," J. Non-Crystalline Solids **121**, 158 (1990).

Weinberg, M.C., Zelinski, B.J., Uhlmann, D.R., and Zanotto, E.D., "On Critical Cooling Rates for Glass Formation," J. Non-Crystalline Solids **123**, 90 (1990).

Weinberg, M.C., "The Quantitative Determination of the Effectiveness of Nucleating Agents," J. Non-Crystalline Solids **122**, 139 (1990).

Weinberg, M.C., "The Effect of Multiple Nucleation Events on Heterogeneous Nucleation Rates," J. Chem. Phys. **92**, 7515 (1990).

6. COMBUSTION SCIENCE

An Experimental and Theoretical Study of Radiative Extinction of Diffusion Flames

Michigan State University
Dr. Arvind Atreya
Dr. Indrek S. Wichman

Objectives: The objective of this project is to perform a detailed investigation of radiation-induced extinction of diffusion flames. Such extinction is not observed under normal gravity conditions due to buoyancy and is expected to occur under μg conditions.

Research Task Description: Radiation-induced extinction is being investigated both experimentally and theoretically. Ground based experiments are being performed to determine the soot particle formation and oxidation rates in a specially constructed low strain rate counterflow diffusion flame apparatus. The results of these experiments will be used in a transient model to predict the radiative extinction limit and the conditions under which it occurs. Simultaneous experiments are being performed under μg conditions at the NASA Lewis Research Center by employing a spherically symmetric diffusion flame to confirm the theoretical predictions.

To examine the radiative extinction limit under μg experiment (2.2 seconds) and because the flame may be radiatively quenched prior to attaining steady state.

Progress to Date: Although this project started in April 1991, we have made considerable progress on the following items:

We are designing a low heat capacity spherical porous burner for the 2.2 sec. drop tower μg experiments. Special care is being taken to ensure that the gas flow from this burner is uniform and that extinction will occur due to radiation and not due to heat loss to the burner.

We have formulated the theoretical problem and are currently examining the time it takes for the diffusion flame to attain steady state. It is important to understand these transient aspects because there is a time constraint on the μg experiment (2.2 seconds) and because the flame may be radiatively quenched prior to attaining steady state.

The experimental and theoretical program for soot formation studies was initiated much before this project was funded. Based on this work we have formulated the soot formation part of the problem. It is now being prepared for publication.

Also a theoretical study of μg droplet burning was begun well before the project was funded. It is also being prepared for publication.

Publications

Wichman, I. S. and Baum, H. R., "A Solution Procedure for Combustion Problems Involving Small Reynolds Number Flows in μg Environments," To be submitted to the 24th Symposium (International) on Combustion, 1992.

Atreya, A. and Chao, Z., "A Model for Soot Formation in Counterflow Diffusion Flames," To be submitted to the 24th Symposium (International) on Combustion, 1992.

Chao, Z., Lee, K. and Atreya, A., "Experimental Correlations of Soot Formation Rates in Counterflow Diffusion Flames," To be submitted to the 24th Symposium (International) on Combustion, 1992.

The Combustion of Free of Unsupported Fuel Droplets at Low Gravity

Cornell University

Professor C. Thomas Avedisian

NAG3-987 (NASA Contact: J. Haggard, LeRC)

October 1989 - October 1990

Objectives: This research seeks to study experimentally the combustion of small unsupported and isolated fuel droplets within a low gravity environment. The emphasis in the experiments has been to examine the burning characteristics of multicomponent droplets, especially droplets which are miscible mixtures of liquids. The experimental method attempts to create a fundamental configuration for the droplet combustion process, namely near spherically symmetric combustion. In this way, the data obtained may be used to evaluate past droplet combustion models which assume spherical symmetry and, if necessary, motivate development of new models. Accounting for asymmetries in droplet combustion which are induced by a significant axial vapor flow around the droplet can complicate model development, especially if it is desired to model other aspects of the droplet burning process such as extinction and soot formation.

Research Task Description: A low gravity environment is created by performing the experiments within a drop tower. The test droplet, its surrounding combustion gases (room temperature air in the present experiments), and associated optical equipment (to record the droplet burning process) are simultaneously released into free-fall. The droplet then experiences a low gravity environment within the moving frame of reference. The photographic results provide information about the occurrence of such phenomena as extinction, microexplosions, and soot formation. The droplets are small enough (nominally with an initial diameter of about 500 μm) that their complete combustion history can be recorded within the available experimental time.

Progress to Date: The primary focus of the research over the past year involved making hardware improvements to enhance data quality, installment of a computerized image analysis system for data reduction from the motion picture movie records of the droplet burning process, and analyses of data using the image analysis system.

Apparatus modifications consisted of: (1) construction of a new drop package; (2) installation of a drag shield around the package; (3) construction of a new ignition circuit to better control spark energy; and (4) construction of a new electrode retraction mechanism to radially retract the electrodes. The new drop package consisted essentially of a laser table to facilitate mounting of instruments. A drag shield was constructed and placed around the package. A drag shield had not been necessary in previous experiments because of the short burning times of the small droplet sizes studied ($\sim .5\text{mm}$ initial diameter): the gravity level (Grashof number) remained low enough during the droplet burning period for buoyancy to exert a negligible influence on the droplet burning process. While the primary focus of this research effort has been on unsupported droplets, over the past year experiments were performed on larger (initial diameter 1mm) suspended droplets to enlarge the data base for burning rates with a view toward exploring a possible effect of initial droplet diameter on burning. These experiments revealed a need for shielding the instrumentation package from the oncoming air throughout the entire period of free fall. Further improvements included the refurbishing of the high-speed movie camera and the mounting of a video camera at 90° to the high-speed movie camera. The video camera aided positioning of the droplet prior to an experiment. A new ignition circuit with controllable current and voltage inputs was built with two sparks created on opposite sides of the droplet. The electrode retraction mechanism was redesigned such that the electrodes retract radially away from the droplet. This retraction minimizes air flow around the droplet due to the electrode retraction because the electrodes have a very small cross sectional area.

A computer based image analysis system (Automatix Inc.®, Billerica, MA) was used to analyze data by magnifying the images from the 16 mm film through a microscope onto a CCD camera which sent the image to the computer. Analyzed data included unsupported droplets of heptane, pure methanol, methanol/toluene mixtures, and methanol/dodecanol mixtures. The frame-by-frame analysis allowed for more clearly specifying extinction diameters for the pure methanol droplets and the methanol/toluene mixture droplets. The multi-staged combustion observed in droplet combustion of some mixtures droplet combustion experiments was definitively brought out for the methanol/dodecanol mixtures with the frame-by-frame analysis. The analyses of the methanol/toluene mixtures showed no such behavior. Results from the computer-based image analysis system were included in the publications listed below.

Publications

Yang, J.C., Jackson, G.S., and Avedisian, C.T., "Combustion of Unsupported Methanol/Dodecanol Mixture Droplets at Low Gravity," *23rd International Symposium on Combustion*, The Combustion Institute, 1991, pp. 1619-1624.

Jackson, G.S., Avedisian, C.T., and Yang, J.C., "Observations of Soot during Droplet Combustion at Low Gravity: Heptane/Monochloro-alkane Mixtures," *Int. J. Heat & Mass Transfer*, 1991 (in press).

Jackson, G.S., Avedisian, C.T., and Yang, J.C., "Soot Formation during Combustion of Unsupported Methanol/Toluene Mixture Droplets in Microgravity," in *Proceedings of the Royal Society of London*, 1991 (in press).

Presentations

Jackson, G.S. and Avedisian, C.T., "Combustion of Multicomponent Droplets in a Microgravity Environment," *Gordon Research Conference on Gravitational Effects in Physico-chemical Systems*, Plymouth, NH, June 1991.

Ignition and Combustion of Bulk Metals in a Microgravity Environment

Center for Combustion Research
University of Colorado at Boulder
Professor Melyvn C. Branch
Professor John W. Daily (Mechanical Engineering)
Professor William B. Krantz (Chemical Engineering)
NAG3-1257
April 1, 1991-October 31, 1994

Objectives: The objective of this research is to develop a coordinated research program to investigate the ignition and combustion characteristics of bulk metals in a microgravity environment. Since metals are known to be combustible, it is important to identify ignition conditions and the effect of the ambient environment in order to identify flame-inhibiting atmospheres for space station use of such materials. The study of metal combustion in a microgravity environment, where gravity-driven natural convection is eliminated, will provide new data for comparison to experiments in the presence of gravity. A major additional benefit of the proposed research is that new experimental capabilities will be developed for use in this and future microgravity combustion research projects.

Research Task Description: Cylindrical specimens of bulk metals will be ignited by radiant heating in an oxygen environment within a microgravity combustion apparatus. Measurements will be made of surface and subsurface temperature with a two wavelength pyrometer and a micro-thermocouple, instantaneous mass will be measured by a new mass-change detector and surface and flame characteristics will be recorded by flame emission and schlieren imaging and spectral analysis. Surface morphology of reacted or arrested specimens will be analyzed by scanning electron microscopy. It is planned to complete measurements in normal gravity, elevated gravity by mounting the experimental apparatus on a geotechnical centrifuge and in the low gravity of a NASA operated spacecraft. The goal of the research is to provide new fundamental data on the combustion mechanisms of the metal specimens and to complete the definition of space flight experiment requirements for experiments on the space shuttle.

Progress to Date: Based on the results obtained from preliminary experiments and motivated by the need to test capability of the different possible radiation sources for metal heating, a first combustion chamber and experimental set-up has been designed and is under construction. The design included a 5-liter cylindrical chamber mounted in the vertical position with a 135mm diameter quartz window on the top cover which will permit both the irradiation and temperature measurement of the sample. The metal specimen will be located in a pedestal attached to the bottom cover. Oxygen will be introduced through a filling/evacuation inlet port for final pressures ranging from sub-atmospheric to 150 psia. The inside pressure will be monitored by a 0-3000 psia pressure transducer and the subsurface temperature from the metal specimen will be measured by a Pt-Pt10%Rh thermocouple. Several Xenon lamps (different type and power) will be used as radiation sources to test their effectiveness in metal ignition. These lamps as well as the infrared pyrometers for temperature measurements will be obtained from the manufacturers as demonstration equipment for a 30-day period. The results from these tests will provide reliable information about the equipment capability which in turn will lead us to the design of an improved experimental apparatus for the high-g and low-g tests.

Modeling of Microgravity Combustion Experiments

University of Illinois, Urbana
Dr. John David Buckmaster

Objectives: To carry out fundamental theoretical studies designed to elucidate a variety of phenomena that have been observed in microgravity combustion experiments. This involves the formulation of mathematical models and their solution. These solutions are to be obtained primarily by analytical means, but numerical tools will be used where appropriate and necessary. The relation between these solutions and the experimental data will be qualitative for the most part, although some quantitative conclusions may be possible. It is expected that the results will lead to: an understanding of the physical origins of the several behaviors; the identification of mathematical models which could provide the foundation for quantitatively accurate numerical simulations; and experimental feedback, that is, the suggestion of new experiments, or the suggestion of new parameter ranges to explore in old experiments.

Research Task Description: Three phenomena have been the focus of the project to date: (i) particle-cloud flames, with particular attention to a pulsating instability revealed by experiments that is responsible for what are called 'chattering flames'; (ii) flame-balls, stationary spherical flames observed in very lean mixtures of small Lewis number; (iii) the behavior of unsteady flames generated by point ignition with particular attention to confinement effects, and the sensitivity of observed behavior to the initial conditions.

The PI has been engaged in a program of microgravity combustion theory since 1989, with support from NASA since Feb. 1991. It would give a misleading impression of the program at Illinois to confine the present description only to that work which has been carried out since Feb. 1991, especially since substantial progress was achieved as part of the proposal generating activity in 1989, and during the post-submission, pre-funding period in 1990. Here then an overall description is provided, and the specific progress made during the last funding period is described later.

Flame balls are observed in very dilute mixtures, close to the flammability limit, when the controlling Lewis number is significantly less than 1. They are a new phenomena and so worthy of study on that account; and there is promise that fundamental understanding of near-limit mixtures and the flammability limit will come from this study. Substantial theoretical understanding of flame-balls has been generated, and is reported in 4 papers. There it is shown that stable, physically realizable flame-balls can only occur if there are heat losses from the flame, typically from radiation. For most of the experiments this would be band radiation from water produced as a combustion byproduct.

A key result is that flame balls can only occur for a strictly defined range of mixture strengths. If the mixture is too weak, the heat losses will quench the flame; if the mixture is too strong a three-dimensional instability will cause the flame ball to fragment.

Heat losses can occur from mechanisms other than radiation, suggesting the possibility of precise experimental control. In KC-135 tests, g-jitter will generate such flows (but without control of course).

A key experimental observation is that flame-balls are only observed if the Lewis number is fairly small. For a Lewis number of 1 there are no stable flame-ball solutions, and stable solutions for a significant range of mixture strengths only occur when the Lewis number is reduced significantly below 1.

Current efforts on this part of the project are devoted to an examination of the three-dimensional unsteady dynamics of deformed flame-balls, since these are observed in some experiments. And to theoretical treatments using more sophisticated models than have hitherto been used in an attempt to draw more precise quantitative conclusions.

Unsteady spherical flames generated by point ignition. Experiments for mixtures with a Lewis number less than 1 (but not so small as to lead to flame-ball formation) reveal two characteristics: a flame-radius that grows initially as the square-root of time; and self-extinguishing flames in which quenching occurs at some finite radius whose value is extraordinarily sensitive to the initial spark energy. Current work is devoted to seeking an understanding of the sensitivity to the spark energy. Preliminary results suggest that our model does not contain the ingredients necessary for such an understanding, but a useful byproduct of the work is a description of the effects of confinement (finite combustion chamber volume).

Instabilities ('chattering flames') are observed for some operating conditions. We believe that this is an acoustic instability driven by the slip (thermal and kinematic) between the two phases. A general theory has been developed to describe instabilities of this kind. Particle-cloud flames provide one of the few (if not the only) configurations in which a rational acoustic stability theory can be constructed in the context of plane flames without external heat sinks (such as wire-gauze inserts). Consequently, they are an attractive vehicle for the furtherance of our understanding of these important instabilities.

Progress to Date: Flame-balls near the three-dimensional stability boundary display a range of complicated unsteady three-dimensional behavior. In some cases, a ball may get stretched out into a long cylinder of flame, a 'flame-string'. Flame-strings display a peristaltic instability. A theory of flame strings and their stability has been developed which agrees, qualitatively, with the experimental observations. Further work along these lines involves a mathematical treatment valid near the neutral stability point, and leads to unsteady ellipsoidal flames. This analysis is nearly complete, and a paper is being prepared, which will be completed Dec.1, 1991.

A general theoretical strategy has been developed for describing acoustic instabilities for particle-cloud flames traveling in tubes. The method can be applied to any flame structure, an important point since there are many uncertainties concerning the structure of the experimental flames. A detailed development has been carried out when the structure is assumed to be that of a classical gaseous premixed flame, except that the fuel is generated by pyrolysis of the particles in a preheat zone controlled by heat conduction. Realistic estimates of the radiation field show that the radiative pre-heating mechanism proposed in ref.[8] is small. A paper is presently being prepared which will be completed by Oct.1, 1991.

In the context of unsteady spherical flames generated by point ignition, we have modified the analysis in two respects. One involves a correction in that we properly account for the flow field generated by radiative cooling of the burned gases, something that is not done in the earlier work. The other is an accounting for the pressure increase generated by the flame confinement in a fixed volume chamber. A complete theoretical development has been completed, and we have a working code for the numerical aspects of the problem, so that we expect to finish a paper on this work by Jan.1, 1992.

Publications

Buckmaster, J., Joulin, G., and Ronney, P., "The Structure and Stability of Nonadiabatic Flame-Balls," Comb. & Flame 79, 381-392 (1990).

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Buckmaster, J., Lee, C.J., Joulin, G., and Ronney, P., "Modeling of Microgravity Ignition Experiments," in *Recent Advances in Combustion Modeling*, Volume 6 (B.Larrourou, ed.), Series in Advances in Mathematics for Applied Sciences, pp.1-18, World Scientific Press, 1991.

Lee, C.J. and Buckmaster, J., "The Structure and Stability of Flame-Balls: a NEF Analysis," Siam J. Appl. Math., 1991 (to appear).

Buckmaster, J., "A Flame String Model and its Stability," Comb. Sci. & Tech., 1991 (to appear).

Gravitational Effects on Premixed Turbulent Flames: Studies of the Dynamics of Wrinkled Laminar Flames in Microgravity

Lawrence Berkeley Laboratory
University of California, Berkeley
Dr. Robert K. Cheng

Objectives: The main objective of the experimental program is to investigate the dynamics of low Reynolds number premixed turbulent flames in microgravity environment. The aerodynamics flow field and mean flame properties of flames subjected to low gravity, +g and -g forces will be characterized by the use of laser based visualization techniques such as schlieren and tomography. The data will be used to elucidate the underlying physics of the effects of gravity on the complex interaction between turbulence and combustion reactions. The results will also be used for comparison with and guide the development of current theoretical turbulent combustion models which generally do not include the effects of gravity. A range of experimental conditions will be explored for the purpose of identifying candidate experiments to be proposed for space based combustion research facilities.

Progress to Date: In preparation for the microgravity experiments which will be conducted in the 2.0 second drop tower at NASA Lewis Research Center, a laser schlieren study of small Bunsen and rod-stabilized v-flames in +g and -g environments has been initiated. The main goal is to determine the range of conditions under which gravity has a significant effect on flame propagation. The apparatus and schlieren system also serve as the prototype for optimizing the burner design, the schlieren system the flow supply and control system for adaptation to the physical confinements of the drop package.

Combustion of Interacting Droplet Arrays in a Microgravity Environment

NASA Lewis Research Center
Daniel L. Dietrich, Sverdrup Technology Inc., LeRC Group
John B. Haggard, Jr., NASA Lewis Research Center
March 1991 to present

Objectives: This research program represents a comprehensive effort to observe and define the behavior of one, and two dimensional arrays of droplets burning in microgravity. The research program is intended to compliment the existing droplet combustion program at NASA and extend it to examine the effects that multiple droplet interactions have on the combustion characteristics of droplets. The data obtained from this study are expected to improve our understanding of spray combustion systems on earth and in space environments and provide fundamental data from which to compare existing and future models.

Research Task Description: The program is primarily experimental and uses droplets supported on small fibers. The primary independent variables in the study are the initial droplet size and inter-droplet spacing, the ambient pressure and oxygen concentration, diluent gas and the fuel type. The measured variables consist of the temporal variation of droplet and flame size and flame shape. The majority of the work will be conducted in the small drop tower at the NASA Lewis Research Center. Later in the program the zero-gravity facility at Lewis and possibly the low gravity aircraft may be used.

Progress to Date: To date, both a normal gravity and low gravity test facility have been specified and ordered. The normal gravity pressure chamber is nearly operational. In addition, a droplet has been successfully dispensed, deployed and ignited on a small 10-15 micron Si-C fiber using one of the droplet combustion rigs at the small drop tower. This is significant, because this fiber is an order of magnitude smaller than the fibers typically used in droplet studies. These fibers may also be used to obtain temperature measurements in droplet burning.

Publications

Friedman, R. and Dietrich, D., "Fire Suppression in Human-Crew Spacecraft," NASA Technical Memorandum 104334, (1991); also presented at the Halon Alternatives Technical Working Conference, Albuquerque, New Mexico, April 30 - May 1, 1991; also to appear in J. Appl. Fire Sci.

Dietrich, D., Ross, H.D., and T'ien, J.S., "Observations of Candle Flames in Low Pressure and Low Gravity," presented at the 1990 meeting of the Eastern States Section of the Combustion Institute, Orlando, Florida.

Scientific Support For A Proposed Space Shuttle Droplet Burning Experiment

Princeton University

Professor Frederick L. Dryer

NAG 3-1081 (NASA Grant: Mr. John B. Haggard, Jr., LeRC)

October 3, 1989 - November 30, 1991

Joint Research With University Of California, San Diego, Professor Forman A. Williams

Objectives: The general objective of this program is to ascertain how best to utilize reduced gravity facilities to pursue scientific investigations of droplet combustion. The scientific objective is to provide scientific support for development of a droplet combustion experiment module for conducting experiments in the NASA LeRC microgravity facilities, and possibly aboard a space-based platform such as the Space Shuttle or in the Space Station. The planned experiments are intended to improve fundamental understanding of droplet combustion phenomena, especially in relationship to time-dependent characteristics and extinction phenomena. The scientific-support objective encompasses data acquisition and reduction as well as asymptotic and numerical modeling for interpretation of observations.

Research Task Description: The research tasks include theoretical modeling using numerical techniques and asymptotic methods, with emphasis on transient phenomena, detailed chemical kinetics and transport effects, ground-based experimentation on droplet burning in both the 2.2 and 5.5 second NASA LeRC droptowers, support to NASA in providing advice on hardware aspects of a flight experiment and analysis of data to be obtained from all experiments. The modeling addresses questions related to transient burning, to soot formation and destruction, to droplet disruption, and to ignition and extinction phenomena. Ground-based experiments have been focussed on droplet ignition and on impulses imparted to droplets by ignition sparks; spark designs for minimum impulse were addressed. In addition, droptower experiments have addressed burning rate measurements and mechanisms of soot production and of droplet disruption, as well as extinction phenomena. Present work encompasses n-decane, n-heptane and methanol as fuels in oxidizer/inert mixtures at pressures between about 0.2 and 2 atmospheres.

Progress To Date: In this work, we have obtained data on the spherically symmetric burning of decane, heptane and methanol droplets in a range of ambient environments. Also, the importance of soot formation and how it influences the burning characteristics of hydrocarbon fuel droplets were demonstrated. Methods to reduce the sooting propensity of alkane fuels utilizing pressure/inert/oxygen index have been studied. The development of methods to employ a digital data processing technique for analyzing droplet combustion data was completed and this was applied to droplet burning data from droptower experiments. These methods have been supplied by Princeton to other investigators for use in similar data processing applications. Combustion-generated product dissolution in the liquid phase of a droplet was studied, and histories of impurity build-up in the liquid during droplet combustion was determined. Utilizing the diluent substitution method, the first well-characterized extinction of a spherically symmetric burning droplet was observed.

A new, transient, spherically symmetric numerical model utilizing detailed chemical kinetics and transport properties was developed and this computer code was utilized to study the time dependent features of droplet burning phenomena. Simultaneously calculated burning rate, flame stand-off ratio and extinction diameters without the use of empirical parameter adjustments agree well with those determined experimentally for methanol droplet burning in oxygen/helium environments. Calculated absorption/dissolution of water and formaldehyde into pure burning methanol droplets were compared with experimental results. Finally, the numerical calculations were utilized to predict experimental test envelopes for microgravity experiments on methanol and n-heptane droplet burning.

Publications

M.Y. Choi, Y.S. Stein and F.L. Dryer, "Initial Investigations On The Combustion Behavior Of Methanol/Water Mixtures," To Be Presented At The 30th Aerospace Sciences Meeting: Sixth Annual Space Processing Conference, Reno, NV., Jan 6-9, 1992.

J.B. Haggard, Jr., B.A. Borowski, F.A. Williams, J.Card, F.L. Dryer and M.Y. Choi, "Microgravity Combustion Of Single, Pure, n-Decane and n-Heptane Droplets In The NASA-Lewis 5 Second Zero-Gravity Facility," To Be Presented At The 30th Aerospace Sciences Meeting: Sixth Annual Space Processing Conference, Reno, NV., Jan 6-9, 1992.

S.Y. Cho, R.A. Yetter and F.L. Dryer, "Computer Model For Chemically Reactive Flow With Complex Chemistry/Multi-Component Molecular Diffusion/Heterogeneous Processes," *J. Comp. Phys.*, 1991 (in press).

M.Y. Choi, S.Y. Cho and F.L. Dryer, "Experimental/Computational Basis For Conducting Droplet Combustion Experiments On Space-Based Platforms," in *Proceedings of IUTAM Symposium On Microgravity Fluid Mechanics*, ZARM University Of Bremen, Reviewed and published in the proceedings.

M.Y. Choi, F.L. Dryer and J.B. Haggard, Jr., "Some Further Observations On Droplet Combustion Characteristics: NASA-Princeton Results," *AIAA/IKI Microgravity Science Symposium*, Moscow, USSR, May 13-17, 1991.

M.Y. Choi, F.L. Dryer and J.B. Haggard, Jr., "Observations Of A Slow Burning Regime For Hydrocarbon Droplets: n-Heptane/Air Results," *Twenty-Third Symposium (International) On Combustion*, The Combustion Institute, 1991, p. 1597.

S.Y. Cho, M.Y. Choi and F.L. Dryer, "Extinction Of A Free Methanol Droplet In Microgravity", *Twenty-Third Symposium (International) On Combustion*, The Combustion Institute, 1991, p. 1611.

J.B. Haggard, Jr., M.H. Brace, F.L. Dryer, M.Y. Choi, F.A. Williams and J.Card, "N-Decane Droplet Combustion Experiments In The NASA-Lewis 5 Second Zero-Gravity Facility: Results In Tests Other Than In Air", AIAA Paper No. 91-0720, 1991.

Combustion of Electrostatic Sprays of Liquid Fuels in Laminar and Turbulent Regimes

Yale University
Dr. Alessandro Gomez
Marshall B. Long
Mitchell D. Smooke

Objective: The project objective is to study the formation and burning of electrosprays of liquid fuels at both normal and reduced gravity in laminar and turbulent regimes. The most distinctive advantages of the electrospray over alternative atomization techniques are: the generation of a narrow droplet size distribution; the self-dispersion property of the spray due to coulombic repulsion; the absence of droplet coalescence; and the opportunity of manipulating the trajectories of charged droplets. We exploit these advantages in some novel applications of the electrospray in the area of multiphase combustion. Practical sprays consist of two-phase flows of a generally polydisperse distribution of droplets in a turbulent gaseous environment. We are planning to synthesize gradually these complex sprays, by using the electrospray to generate controlled clouds of droplets. Since liquid atomization mechanisms, the host gas where the liquid is injected can be controlled independently. The underlying philosophical approach is that of providing an experimental arrangement that would allow a systematic study of spray evolution and burning in configurations of successively increasing level of complexity, starting from laminar sprays to fully turbulent ones. This approach parallels similar efforts made by other research groups in the numerical modeling of sprays, thereby providing an experimental base to test and further develop necessarily approximate theories.

Research Task Description: The first phase (approximately, two years) of the proposed research would be conducted at normal gravity in the Combustion Laboratory at Yale. It would entail: a) the investigation of the mechanism of electrospray atomization and dispersion, with the goal of identifying and characterizing the domain of operating variables under which the generated droplets are monodispersed; b) the experimental investigation on the spray combustion in a counterflow spray burner and c) the numerical modeling of the latter. A second phase of the experimental program (third year) would be partly implemented at the 2s Drop Tower NASA facility, as part of a flight definition program for subsequent Space Shuttle experiments. The goal is that of studying the same types of sprays in an environment free of natural convection effects. From a computational viewpoint, we will develop counterflow spray models by coupling the liquid/gas phase system in such a fashion that the complete problem can be solved with modified version of previously developed algorithms. To investigate droplet/droplet interactions we propose to model droplets in a two dimensional array with detailed transport and finite rate chemistry. The interaction between experiments and models will help in interpreting the experimental results and in validating the computational model. The last phase of the project (fourth year) will be partly devoted to turbulent sprays, in which turbulence is "injected" in the gaseous stream by synthesizing various turbulent scenarios, characterized by different intensities and scales.

Progress to Date: The first seven months of work have been focused on two main tasks: i) the investigation of the mechanism of electrospray atomization and dispersion, with the goal of identifying and characterizing the domain of operating variables under which the generated droplets are quasi-monodisperse and ii) the construction and testing of a laminar spray combustion prototype.

Research on the fundamentals of electrospray atomization is crucial to explore the range of operating variable over which the electrospray can generate monodisperse droplets. A map of the electrospray stability domain in the applied-voltage vs liquid flow rate plane was obtained for heptane, doped with an antistatic additive to provide electric conductivity. A domain was identified in which monodisperse droplets were generated whose size was primarily controlled by the liquid flow rate and, to a less extent, by the applied voltage. For all measurements the droplet standard deviation was less than 10% of the average diameter, the latter spanning a remarkable range of over two orders of magnitude (1-140 μ m). The boundaries of this stability region were determined by the onset of different types of instabilities: at a given flow rate, the minimum voltage was set by the onset of pulsations in the liquid meniscus, whereas the maximum voltage was set by the onset of traverse

instabilities on the jet issuing from the conical meniscus; at a given voltage, the upper limit in flow rate was determined by the onset coulomb explosion, which is achieved when coulomb repulsion of the electrical charges on the droplet surface overcomes surface tension and the droplets disrupts. Indeed, to the best of our knowledge, for the first time Coulomb explosion has been photographically captured. Explosion occurs by the formation of a cone, similar to the stable conical meniscus in the electrospray, which ejects a stream of droplets one order of magnitude smaller in size. Another aspect of electrospray atomization which we are pursuing is the interpretation of droplet size and velocity measurements in a prototypical spray, a 10 cc/hr heptane electrospray, to understand what governs the spray behavior. We have determined the net electric field acting on droplets. We are now in the process of assessing what fraction of this field is due to the external field caused by space charge effects. Assessment of the role of space charge effects is relevant to the determination of factors governing and possibly limiting electrospray performance and is also crucial to future strategies of droplet trajectory manipulation via electrostatic focusing. Results obtained to date were in part presented at ICLASS (International Conference on Liquid Atomization and Spray Systems) in July. A forthcoming presentation will be given at the 10th Annual AAAR (American Association for Aerosol Science) Meeting next week. Three archive publications are being prepared for journal submission within the next two months.

Whereas the experimental set-up to produce monodisperse droplets in an electrospray is extremely simple, its adaptation to combustion environments is nontrivial for the following reasons: first, the electrospray cannot be directly ignited because the combustion region is characterized by relatively large concentrations of chemi-ions and the corresponding environment has an electric breakdown threshold lower than the minimum field necessary to establish a stable spray. We electrically insulated the combustion region from the spray to sandwiching the flame between grounded gauzes, thereby burning in the absence of any external electric field. Second, droplets ejected from the capillary needle at velocities in the order of 10m/s need to be slowed down for the establishment of a stable flame. Failure to do so results, as we observed in preliminary tests, in droplets overshooting the flame and penetrating into the oxidizer region, thereby creating a complex partially diffusion-controlled, partially premixed environment. Consequently, provisions were made for droplet deceleration towards terminal velocity by providing a nearly zero field region in which they could travel for a time comparable to size-dependent particle relaxation time. These are the two main considerations that guided the design of a counterflow diffusion flame burner in which we successfully stabilized a spray flame. Droplet size and axial velocity distribution were measured throughout the spray by a commercial Phase Doppler Anemometer (PDA) (Dantec Electronics). Results indicated that narrow size distribution are injected in the combustion region. Preliminary results will be discussed in the forthcoming Technical Meeting of the Eastern States Section of the Combustion Institute (ESCI) in two weeks.

Publications

Gomez, A. and Tang, K., Proceedings of the Fifth International Conference on Liquid Atomization and Spray Systems, ICLASS-91, Gaithersburg, MD, U.S.A., H.G. Semerjian Ed., NIST Special Publication No. 813, p.805, 1991a.

Gomez, A. and Tang, K., Proceedings of the Fifth International Conference on Liquid Atomization and Spray Systems, ICLASS-91, Gaithersburg, MD, U.S.A., H.G. Semerjian Ed., NIST Special Publication No. 813, p.771, 1991b.

Gomez, A. and Tang, K., "The Electrospray as a Source of Quasi-Monodisperse Droplets in the Supramicron Range", AAAR 10th Annual meeting, Traverse City, MI, October 7-11, 1991.

Chen, G. and Gomez, A., "Burning of a Quasi-Monodisperse Heptane Electrospray in a Counterflow Diffusion Flame", Eastern States Section of The Combustion Institute, Fall Technical Meeting, Ithaca, NY, October 14-17, 1991.

Time-Dependent Computational Studies of Premixed Flames in Microgravity

Naval Research Laboratory
Dr. K. Kailasanath
Dr. Gopal Patnaik
Dr. Elaine Oran
(NASA Contact: Dr. Howard Ross, LeRC)
October 1990 - March 1991

Objectives: The objective of this research is to investigate fundamental problems in the combustion of premixed gases such as the differences in the propagation and extinction of gas-phase flames in zero and normal gravity.

Research Task Description: We study the dynamics of premixed flames by performing detailed numerical simulations using a time-dependent, two-dimensional numerical model. This model solves the multispecies coupled partial differential reactive flow equations. The model includes detailed chemical kinetics mechanisms, algorithms for thermal conduction, molecular diffusion, viscosity, heat conduction to walls, convective transport and effects of gravity. We also evaluate various simplified models by comparing their predictions to those from detailed simulations.

Progress to Date: The emphasis of this year's research effort has been to study the dynamics of flames near the limits of propagation. Flames near the lean and rich flammability limit were investigated. The effects of viscosity and heat losses to the walls on structure of flames near the lean limit of hydrogen were simulated. The simulations of flames in zero gravity show that wall heat losses can promote the formation of multiple cells. The effects of viscous losses to the walls creates dramatic changes in the structures of flames confined within adiabatic walls. However, the effects of viscosity are found to be secondary and can be neglected when more realistic non-adiabatic walls are used. Heat losses are found to significantly modify upward- and downward-propagating flames and many features observed in experiments have also been observed in the simulations. The details of the extinguishment process of downward-propagating flames have been studied and are currently being prepared for publication.

In another series of simulations, the dynamics of flames near the rich limit of hydrogen in air was investigated using the detailed one-dimensional model. The one-dimensional rather than the two-dimensional model was used because the flames in rich hydrogen-air mixtures are nearly planar. These simulations showed that as the rich-limit is approached, a chemical instability occurs which causes the burning velocity to oscillate. The instability mechanism was identified to be due to a competition between certain chain-branching and chain-terminating reactions. Flame extinction itself is unsteady and cannot be studied using steady-state assumptions.

Currently, preparations are being made to study hydrocarbon flames and to investigate the effects of radiative losses on the observed dynamics of flames.

Publications

Patnaik, G. and Kailasanath, K., "Effect of Gravity on the Stability and Structure of Lean Hydrogen-Air Flames", in *Proceedings of the 23rd (International) Symposium on Combustion*, The Combustion Institute, Pittsburgh, PA., 1991, pp. 1641-1646.

Patnaik, G. and Kailasanath, K., "Effect of Gravity on Flame Instabilities in Premixed Gases," AIAA Journal, 1991 (to appear).

Kailasanath, K., "Laminar Flames in Premixed Gases," Prog. Astro. Aero. **135**, 225-256 (1991).

Presentations

Patnaik, G. and Kailasanath, K., "Effect of Heat Losses on the Cellular Structure of Lean Hydrogen-Air Flames," *29th Aerospace Science Meeting*, Reno, January 1991, AIAA Paper 91-0784.

Kailasanath, K., Ganguly, K., and Patnaik, G., "Dynamics of Flames near the Rich-Flammability Limit of Hydrogen-Air Mixtures," *13th International Colloquium on the Dynamics of Explosions and Reactive Systems*, Nagoya, Japan, July 1991.

Radiative Ignition and Subsequent Flame Spreading in a Microgravity Environment

National Institute of Standards and Technology

Dr. Takashi Kashiwagi

Dr. Howard R. Baum

Objectives: The objective is to develop theoretical models capable of predicting ignition and subsequent flame spreading over a cellulosic material in a microgravity environment using the material characteristics determined in normal gravity. This study is to lead to applications such as fire safety for spacecraft by understanding and predicting ignition and subsequent flame spreading behavior.

Research Task Description: This study consists of two parts: (1) An experimental study in normal gravity is conducted to determine degradation characteristics of a cellulosic material and kinetic constants of global gas phase oxidation reaction of the evolved degradation products. Global kinetic constants of thermal and oxidative degradation reactions are measured by using a thermal analysis technique. (2) Theoretical models on ignition and subsequent flame spread are developed and solved numerically using the measured characteristics of the degradation and of the global gas phase oxidation reactions. The advantage of a microgravity environment is fully utilized, the gas phase flow pattern is calculated by solving an irrotational flow pattern at a low Reynolds number regime. The calculation is conducted in two-dimensional, axisymmetric, and three-dimensional configurations. A forced slow flow up to 10 cm/s (simulating a ventilation flow in a spacecraft) is included in the calculation to determine the effects of the wind on the transition from ignition to flame spread. The majority of the calculation is conducted with a thermally thin material but limited results are available for a thermally thick material.

Progress to Date: Theoretical models and numerical codes for spontaneous ignition and subsequent flame spread have been developed in the two-dimensional configuration with a slow wind and also in a quiescent axisymmetric configuration. The gas phase model is based on irrotational flow mainly controlled by the slow forced flow, gas expansion and mass addition from a degrading material with one-step oxidative reaction with energy and chemical species equations. The condensed phase model is based on the thermally thin cellulosic sheet with three global degradation reactions, pyrolysis reaction and oxidative degradation of the sheet to generate char and gaseous products, and highly exothermic oxidative char degradation. Preliminary results show that ignition parameters, the size of irradiated area, its duration and flux, have significant effects on transition to flame spread. It appears that transition from spontaneous ignition to flame spread tends to be difficult without the slow external flows. A three-dimensional code without gas phase oxidation reactions has been developed. A heat transfer problem with a thermally thick sample has been studied. Determination of global kinetic constants of the three degradation reactions was completed using TGA analysis with multiple heating rates and oxygen concentrations and by continuous evolved gas analysis of CO, CO₂, H₂O and O₂.

Publications

Kushida, G., Baum, H.R., Kashiwagi, T. and Di Blasi, C., "Heat and Mass Transport From Thermally Degrading Thin Cellulosic Materials in a Microgravity Environment", accepted publication in J. Heat Transfer.

Kashiwagi, T. and Nambu, H., "Global Kinetic Constants for Thermally Oxidative Degradation of a Cellulosic Paper", submitted in Combustion and Flame.

Yamashita, H., Baum, H.R. and Kashiwagi, T., "Heat and Mass Transport From Heated Material in a Low Reynolds Number Microgravity Environment", to be presented at Fourth International Conference on Numerical Combustion sponsored by SIAM, December 1991.

Measurements and Modeling of Sooting Turbulent Jet Diffusion Flames under Normal and Reduced Gravity Conditions

Wayne State University
Dr. Jerry Ku

Progress to Date: Derivation of a solution for radiative heat transfer in axisymmetric, finite cylindrical enclosures, based on the P3 spherical harmonics approximation and a scattering phase function represented by a series of Legendre polynomials, has been completed. This is different from that derived by Menguc and Viskanta (J. Heat Transfer 108, 271-276, 1986), who used a phase function represented by the delta-Eddington approximation for highly forward scattering cases. The latter is not applicable to this study, since we have concluded that soot agglomerates do not have a highly forward scattering pattern. We are now in the process of programming the solution. An elliptic differential equation solver, ELLPACK (Solving Elliptic Problems using ELLPACK, Rice and Boisvert, Springer-Verlag, 1984), needed for this work, has been ordered from Purdue University.

Soot formation model developed by Syed et al. (23rd Int'l Symp. Comb., 1533-1541, 1990) has been successfully incorporated into J-Y. Chen's (18th Annual Pittsburgh Conf. on Modeling and Simulation) block-tridiagonal parabolic solver code for nonpremixed combustion turbulent free-shear flows. We are now in the code-verification process - comparing our results against other numerical results.

A piloted diffusion flame burner nozzle has been designed and fabricated. Testing will be started soon.

Studies of Flame Structure in Microgravity

Princeton University
Professor Chung K. Law

Objectives: The general objectives of this work are to study the response and structure of flames in simple, well controlled flow fields and compare with theoretical and numerical results to provide increased understanding of flame processes. The flames to be investigated are cylindrical and spherical flames under microgravity conditions. Specific objectives are to determine the flame stabilization mechanism, either through heat loss or flow divergence, study the effects of flame curvature on flame response, and investigate thermo-diffusive flame front instability.

Research Task Description: Experiments will be performed in the ground-based microgravity drop tower at NASA Lewis. Cylindrical and spherical porous burners will be used to provide well controlled divergent flow fields for the flames. Initial data will be obtained using conventional thermocouple measurements and photographic techniques, while later measurements will be done using laser-based techniques that are currently being developed at NASA. Measurements of interest are the mass burning rate, burner surface temperature, and flame front standoff distance. The experimental data will be compared with both analytical results and numerical simulations. Specific issues of interest in analytic studies are the flamefront instability for stationary flames in the presence of flame curvature but without flame stretch. The use of one-dimensional flame codes using the latest kinetic models will allow quantitative comparisons with the experiments and further substantiate the experimental measurements

Progress to Date: The existing combustion chamber has been modified to accommodate our experimental requirements, and cylindrical porous burners have been made. Initial experiments have been performed at the drop tower to determine the qualitative response of cylindrical flames in microgravity, and to resolve any difficulties that exist with the experimental apparatus. The premixed flame code from Sandia has been modified for use in the cylindrical geometry and initial numerical results have been obtained.

Publications

Eng, J.A., Egolfopoulos, F.N., and Law, C.K., "On the Dual Response and Surface Recombination in Burner-Stabilized Flames," presented at 28th National Heat Transfer Conference, Minneapolis, MN; Heat Transfer in Fire and Combustion Systems, HTD-Vol. 166, 35, 1991.

A Fundamental Study of the Combustion Synthesis of Ceramic-Metal Composite Materials Under Microgravity Conditions

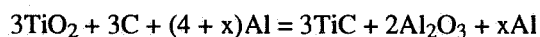
Colorado School of Mines

Dr. John J. Moore

Dr. D. G. Wirth, Coors Ceramic Company

Dr. J. Birch Holt, Lawrence Livermore National Laboratory

Objectives: The objectives of the research program are concerned with obtaining a thorough scientific understanding of the effect of microgravity on the in situ synthesis of ceramic-metal composite materials using an innovative mode of combustion synthesis, e.g.,



The combustion synthesis reaction provides a highly porous product. This research program investigates the use of excess aluminum ($x\text{Al}$) which becomes liquid at the ignition temperature of the exothermic combustion synthesis reaction and is allowed to simultaneously infiltrate the porous ceramic ($\text{TiC-Al}_2\text{O}_3$) matrix.

Several ceramic-metal composite systems will be studied using this approach and various process parameters will be varied in order to determine their effect on the synthesis efficiency and control of morphology of the phases of the ceramic-metal composite produced.

Research Task Description: The initial research that was conducted in this program showed that the liquid aluminum infiltrated the porous ceramic matrix but was concentrated in the lower half of the composite due to gravity driven fluid flow. The main theme of this research program will be to conduct these experiments in microgravity in order to provide a more uniform flow of the liquid aluminum within the porous ceramic matrix.

Initially, the research program will be concerned with conducting these experiments in normal gravity conditions in order to determine the process parameters that should be controlled under microgravity conditions. Subsequently, the most promising experimental conditions will be used to determine the effect of microgravity on these combustion synthesis reaction systems.

Progress to Date: This research program was funded by NASA from August 1, 1991. However, some initial exploratory work was conducted in-house within the Department of Metallurgical and Materials Engineering at Colorado School of Mines during the previous year. This work was concerned with the effect of process parameter, e.g., particle size, green density, reaction stoichiometry, on the synthesis efficiency and stability of the combustion reaction. This area of examination is still in progress and will be completed within the next six months.

Publications

Feng, H.J., Moore, J.J., and Wirth, D.G., "Combustion Synthesis of Ceramic-Metal Composite Materials: The $\text{TiC-Al}_2\text{O}_3\text{-Al}$ System," Met. Trans., 1991 (to be published).

Feng, H.J., Moore, J.J., and Wirth, D.G., "Combustion Synthesis of Ceramic-Metal Composite Materials: The $\text{ZrB}_2\text{-Al}_2\text{O}_3\text{-Al}$ System," *First International Conference on SHS of Materials*, USSR Academy of Sciences, Alma-Ata, USSR, Oct. (1991).

Effect of Low Velocity Forced Flow on Flame Spread Over a Thermally-Thin Fuel in the Absence of Buoyancy-Induced Flows

NASA Lewis Research Center
Sandra L. Olson
In-House*

Objectives: The objective of this program is to determine the extinction limits and steady burning characteristics of thermally-thin solid fuels over a wide range of oxidizer concentrations and flow velocities. This work will provide insight into the effects of buoyancy on flame spread mechanisms by decoupling natural convective transport processes from forced convective, conductive, and diffusive transport processes that are important in spreading flames. Understanding the mechanisms for flame spread will provide a fundamental link between normal gravity materials flammability and materials flammability in spacecraft environments where low velocity forced flows exist due to environmental control systems.

Research Task Description: The approach used in this study is to perform a series of experiments in low gravity varying fuel thickness, oxidizer concentration, diluent composition, pressure, and opposed flow velocity. Tests are conducted in a low speed combustion tunnel developed for use in all three NASA Lewis Research Center's low gravity facilities. Low gravity is required for these experiments because in normal gravity buoyancy-induced gas flows around the spreading flame are on the order of or greater than the range of forced flow velocities to be studied (0-30 cm/s). These natural convective flows overwhelm or combine with the forced convective flows so that the effect of the forced flow on the flame spread rate cannot be isolated.

Progress to Date: Results to date include quiescent flame spread and extinction results which defined an extinction boundary where quenching and blowoff extinction boundaries meet at a low oxygen limit for flame spread in opposed flow. Forced flow flame spread experiments at three different oxygen concentrations show that flame spread rate increases dramatically with opposed flow from the quenching (low velocity) extinction limit to a peak flame spread rate at intermediate opposed flow velocities of 15-20 cm/s. For higher opposed flow velocities, the flame spread decreases until blowoff (high-velocity) extinction occurs. At high enough ambient oxygen concentrations (30% by volume for fuel studied) a wide plateau is observed in the flame spread rate - opposed flow curve where the flame spread rate is independent of opposed flow over a wide range of intermediate flow velocities.

Based upon these results, a flame spread map has been constructed as a function of ambient oxygen concentration and opposed flow velocity which indicates three distinct regions where different mechanisms control the flame spread process. In the near-quenching region (at very low opposed-flow velocities and/or low oxygen concentrations), a new controlling mechanisms of flame spread - oxidizer transport-limited chemical reaction - is proposed. In the near-blowoff region, high opposed flow velocities impose residence time limitations on the flame spread process. A critical characteristic relative velocity line between the two near-limit regions defines conditions which result in maximum flammability both in terms of peak flame spread rate and minimum oxygen concentration for stable burning. In the third region, away from the extinction boundary, flame spread is controlled by gas-phase conduction, and is adequately described by thermal theories of flame spread.

The new mechanism of oxygen transport-limited flame spread has been proposed for the near-limit low velocity flame spread region, which is unique to low gravity environments. It is suggested that the transport-limited heat generation rates, when compared to the significant heat loss rates, control the flame spread process in this region, and heat generation/heat loss ratio reaches a critical value at the quenching extinction limit.

Microgravity quiescent flame spread experiments in various oxidizer-diluent atmospheres indicate that the primary effect of the diluent gas in the near-quenching region is to modify the heat losses from the flame. This changes the critical ratio of heat generation to heat loss and thus shifts the extinction limit. The use of carbon dioxide as diluent (which has strong gas-phase radiative characteristics and a higher heat capacity, both of

which are net heat losses for these weak flames) causes flame extinguishment at much higher oxygen concentrations than nitrogen. Helium, due to its high thermal diffusivity, has high conductive heat losses and despite its lower heat capacity, also extinguishes flames at higher oxygen concentrations than nitrogen. Argon, which has the same thermal diffusivity as nitrogen, causes the flames to extinguish at lower oxygen concentrations because of its lower heat capacity and lower thermal conductivity, both of which reduce heat losses from the flame. Preparation of a manuscript of the diluent extinction results is underway.

An analysis of thickness effects has suggested that the classical definitions of thermally-thick and thermally-thin fuels may not be appropriate in low gravity, where the flame spread process is no longer controlled by conduction either in the gas phase for thermally-thin fuels, or in the solid for thermally-thick ones. Rather, the effect of increasing fuel thickness in low gravity may increase the heat loss due to in-depth solid phase conduction, thus changing the critical ratio between transport-limited heat generation to heat losses. Further analysis and ground-based experiments will be conducted in opposed flow flame spread to better define the critical ratio between transport-limited heat generation to heat losses.

A concept for a flight experiment on "Diffusive and Radiative Transport in Fires (DARTFire)" has been defined by R.A. Altenkirch, S.L. Olson, and S. Bhattacharjee. The DARTFire Experiment will examine the importance of mass diffusion and thermal radiation on the flame spread and extinction mechanisms in low gravity. A draft Science Requirements Document for DARTFire has been written, and a Conceptual Design Review is planned for November-December 1991.

* This work has progressed from ground-based to flight experiment status.

Publications

Olson, S.L., "Mechanisms of Microgravity Flame Spread Over a Thin Solid Fuel: Oxygen and Opposed Flow Effects," Combustion Science and Technology **76**, 233-249 (1991).

Bhattacharjee, S., Altenkirch, R.A., Olson, S.L., and Sotos, R.G., "Heat Transfer to a Thin Solid Combustible in Flame Spreading at Microgravity," Transactions of the ASME: Journal of Heat Transfer **113**, 670-676 (1991).

Presentations

Olson, S.L., "Fuel Thickness Effects on Flame Spread and Extinction Limits in Low Gravity as Compared to Normal Gravity," to be presented at the Eastern States Section of the Combustion Institute, Ithaca, N.Y., October 14-16, 1991.

The Structure of Particle Cloud Premixed Flames

University of California, San Diego
Professor K. Seshadri
Professor A.L. Berlad

Objectives: The principal objective of this research is to clarify the mechanism of propagation of flames in combustible mixtures of fuel particulates and air.

Research Task Description: A numerical and analytical study of the structure and mechanism of propagation of flames in clouds of particulates are planned. The governing equations of mass, and energy, will be integrated numerically to determine the burning velocity of flame. The results will be used to develop an asymptotic analysis of the structure of the flame. In addition it is planned to examine in detail the influence of radiative transport on the structure of the flame.

Progress to Date: The progress to date are summarized in the publications presented below.

Publications

Berlad, A.L. and Tangirala, V., "Particle Segregation Effects," Progress in Astron. & Aeron. **132**, 59-72 (1991).

Berlad, A.L., Tangirala, V., and Seshadri, K., "Particle Nonuniformity Effects on Particle Cloud Flames in Low Gravity," AIAA Paper 91-0718, *29th Aerospace Science Meeting*, Reno, 1991.

Berlad, A.L., "Combustion of Clouds of Volatile Fuel Particulates, Invited Review," Progress in Energy and Combustion Science, 1991 (to appear).

Seshadri, K., Berlad, A.L., and Tangirala, V., "The Structure of Premixed Particle-Cloud Flames," Comb. & Flame, 1991 (submitted).

Combustion Experiments in Reduced Gravity with Two-Component Miscible Droplets

University of California, Davis
Professor Benjamin D. Shaw
March 15, 1991 - September 30, 1991

Objectives: The objective of this research is to study the combustion characteristics of individual and unsupported two-component miscible droplets in reduced-gravity environments. Attention is focused upon droplets that are initially in the millimeter size range. Droplet components are selected so that their volatilities are significantly different. Some phenomena of interest are transient gas-phase processes (e.g., sudden flame contraction caused by rapid increases of the surface mass fraction of the less volatile component, and extinction), transient liquid-phase processes (e.g., droplet diameter histories, bubble nucleation, and disruption), and sooting and transport of observable soot particles (from thermophoretic and aerodynamic forces).

Research Task Description: Experiments will be carried out in the NASA LeRC 2.2 and 5 sec drop towers. Data will be obtained by cine photography of the burning droplets. Films will be analyzed both manually and with digital image processing techniques. Time histories of droplet and flame diameters and other observables will be measured. In conjunction with the experiments, theoretical studies will be undertaken to aid interpretation of the experimental results. An objective of these studies will be to clarify the influence of liquid-phase species and energy transport on transient combustion behaviors (e.g., sudden flame contraction).

Progress to Date: None.

Ignition Delay and Flame Spread Above a Liquid Fuel Pool

University of California, Irvine
Professor William A. Sirignano
Dr. D. N. Schiller
Dr. L. Chao

Objectives: This study is intended to investigate the relative importance of buoyancy and surface tension on ignition delay and flame spread above an axisymmetric liquid fuel pool. By studying the heat and mass transport mechanisms which control ignition delay and flame spread rates, we hope to gain knowledge which may be used to prevent liquid fuel pool fires. When a liquid fuel is initially below its flash point temperature, the liquid fuel must be heated sufficiently to create a combustible mixture of fuel vapor before ignition and flame spread can occur. Buoyancy and thermocapillary forces affect the heating of the liquid pool, the transport of fuel vapor to the ignition source, and the mixing of fuel vapor and air in front of the flame leading edge.

On account of nonuniform heating from above due to the ignition source or the flame, fluid motion will be driven by both surface-tension gradients and by buoyancy. If only liquid motion were considered, then since these driving forces generally support in concert surface fluid motion away from the heat source, one would be led to believe that increasing either buoyancy or surface tension would delay ignition yet support flame spread (as the convection assists the preheating process ahead of the flame). However, heat and mass transport in the gas phase may alter this conclusion. For example, increasing gas-phase buoyancy decreases the heat flux to the liquid surface (since convection toward the ignition source opposes conduction toward the liquid surface), but increases the rate of transport of fuel vapor from the liquid surface to the ignition source. One objective of this research program is to study the relative importance of liquid heating and gas-phase mass transport to ignition delay.

It is well known that when the initial liquid pool temperature is well below the flash point temperature, the flame periodically accelerates and decelerates as it alternates between premixed and diffusive burning. The reason why flame spread alternates between premixed and diffusive burning, however, has not been clearly identified. The present study is intended to investigate the flame pulsation mechanism for both hydrocarbon and alcohol fuels.

In addition to the axisymmetric problem, we are continuing to study associated three-dimensional phenomena including the effects of g-jitter and liquid surface curvature. These are especially relevant in low-gravity environments in which future experiments might be performed.

Research Task Description: Numerical modelling is used to investigate the aforementioned problems. At the NASA Lewis Research Center, experiments are performed in both normal gravity and reduced gravity environments in order to validate the numerical models and to add insight into the problems. The fundamental aspects of the axisymmetric study have been investigated in various enclosed and open pool configurations. Parameters which have been varied include gravity level, surface-tension coefficient, radiation, geometry and temperature of the ignition source, height of the liquid, and properties of the liquid.

Progress to Date: Results of the axisymmetric problem without the effects of chemical kinetics (i.e., studies of the heat and mass transport in the preignition processes) will soon be published by the Journal of Thermophysics and Heat Transfer (Schiller and Sirignano, 1991). In Part I of this two-part paper, the coupling of buoyancy and thermocapillarity was studied in the liquid phase. In Part II, the transport of heat and mass in the gas phase was suggested to be an important factor in ignition delay. Numerical results using an open pool geometry were presented at the 29th AIAA Aerospace Sciences Meeting (Schiller and Sirignano, Reno, Nevada, January 7-11, 1991).

Most of the computational effort in the past year was directed at studying the effects of finite-rate chemical kinetics on ignition delay and flame spread. The results were presented recently at the 1991 Fall Meeting of the Western States Section of the Combustion Institute (Schiller and Sirignano, Los Angeles,

California, October 14-15, 1991), and will soon be submitted for archival publication. Pulsating flame spread is shown to be caused by a recirculation cell which forms in front of the flame leading edge. This recirculation cell is formed by a combination of opposed flow in the gas phase due to buoyancy and concurrent flow in the liquid phase due to thermocapillary forces. Under microgravity conditions, the recirculation cell does not form and, therefore, pulsating flame spread does not occur. For the first time, hot gas expansion is shown to play a significant role in the flame pulsation process by causing the periodic destruction of the gas-phase recirculation zone.

The computational results obtained over the past five years were incorporated into a doctoral dissertation (Schiller, 1991). This work contains preignition and postignition results using both enclosed and open pool configurations. The majority of the modelling to date has been for n-decane fuel. In the future, alcohol fuels such as methanol will be modelled to compare with widely available experimental results. While it has been observed experimentally that flame pulsations with alcohols are different than with hydrocarbon fuels, no explanation has been given as to why flame pulsations differ with fuel type. For example, with hydrocarbon fuels, a "precursor" flame is observed to pulsate forward and backward in front of the main diffusion flame. However, with alcohol fuels, no backward motion of the flame is observed, i.e., the flame accelerates and decelerates rapidly, but always moves forward. Moreover, the whole flame structure pulsates for simple alcohol fuels, whereas for hydrocarbons such as n-decane, flame pulsations consist primarily of the forward and backward movement of the precursor flame.

Publications

Schiller, D. N., "Combustion Above Liquid Fuel Pools: Buoyant and Surface-Tension-Driven Flow Computations," Ph.D. dissertation, U. C. Irvine, October 1991.

Schiller, D. N. and Sirignano, W. A., "Buoyant-Thermocapillary Flow With Nonuniform Supra-Heating: I. Liquid-Phase Behavior," J. Thermophysics and Heat Transfer 6, 1992 (to appear).

Schiller, D. N. and Sirignano, W. A., "Buoyant-Thermocapillary Flow With Nonuniform Supra-Heating: II. Two-Phase Behavior," J. Thermophysics and Heat Transfer 6, 1992 (to appear).

Schiller, D. N. and Sirignano, W. A., "Ignition and Flame Spread Above Liquid Fuel Pools," to be presented at the 1991 Fall Meeting of the Western States Section of the Combustion Institute, October 1991.

Schiller, D. N. and Sirignano, W. A., "Ignition Delay of a Gas Mixture Above a Liquid Fuel Pool," Paper No. 91-0717, presented at the 29th AIAA Aerospace Sciences Meeting, Reno, Nevada, January 1991.

Tsau, F. H., Elghobashi, S. E., and Sirignano, W. A., "Effects of G-Jitter on a Thermally Buoyant Flow," J. Thermophysics and Heat Transfer (accepted).

Combustion of Solid Fuel in Very Low-Speed Oxygen Streams

Case Western Reserve University
Professor James S. T'ien
Kurt R. Sacksteder, NASA/LeRC
NAG3-1046

Objectives: The objective of this research is to study the fundamental characteristics of flame spreading and extinction processes of solid fuels in very low speed concurrent flow.

Research Task Description: The task includes performing both experiments and theoretical modeling work for concurrent-flow flame spread over solid fuel. The flow will be in the low speed regime unobtainable in normal gravity buoyant conditions. Both low speed forced flow and buoyant flow in reduced gravity will be included.

Progress to Date: During the past year, progress was made on two aspects of the experimental program. A series of tests in forced convective, concurrent flow was completed covering a range of 1-5 cm/sec in velocity and 13-30% Oxygen, all at one atmosphere of pressure. Analogous to similar tests in opposed flows, the spreadrates observed increased with the concurrent flow and with the ambient concentration of oxygen. Notably, the test material, a cellulosic laboratory tissue, has been burnt in convecting environments at oxygen concentrations of 13%. To reduce variability in the test results, a significant effort is being made to control the ignition energy applied to the test specimens before the test matrix is expanded to higher concurrent flow velocities.

Secondly, an entirely new testing apparatus has been designed and fabricated for testing buoyant concurrent flows in aircraft tests. The apparatus, currently in the buildup and integration stage, represents the more challenging phase of the experimental program wherein variations in local acceleration will be correlated with flame spreading and flammability. To capture these phenomena, a SAMS transducer will be used for acceleration measurements, and advanced signal conditioning and analysis techniques will be used to clarify the aircraft acceleration environment. A color schlieren system will be used to augment the flame imaging schemes to improve the detection of dim near-limit flames.

In the modeling effort, the objective is to examine the characteristics of concurrent flow flame spread. In this case, flame spread depends on the entire flame (and the associated hot thermal plume, which is important since it preheats the solid fuel) so the computational domain must be relatively large. This is more difficult than opposed flow flame spread, which can be modelled by considering only the leading edge region of the flame.

The flames are modeled using the steady, fully compressible Navier-Stokes equations for the gas phase, together with the conservation of energy and species equations for the gas and solid phases. Since the entire flame must be modelled, and complicated by the fact that these flames tend to be longer than opposed flow flames, use must be made of simplified equations where appropriate. Specifically, when the Reynolds number is big enough, the governing equations become parabolic in nature, enabling much faster computation. Thus, the elliptic equations are solved in the leading edge region (low Reynolds number) and parabolic equations can then capture the long flame tip and thermal plume.

The existing computer algorithm for the elliptic region has been modified to improve performance. The change made enables the pressure field to reach its converged state more quickly. Additionally, the governing equations and corresponding computer code have been formulated for the parabolic (i.e., large Reynolds number) region of the domain. Steady solutions have been obtained with solid phase radiation and forced flow alone (no buoyancy).

The choice of where to begin using the simpler parabolic equations in the domain is still not determined. Ideally, we would like to start at as small a Reynolds number as possible, but of course we want to maintain accuracy, so the starting Reynolds number cannot be arbitrarily small.

Results indicate that flame spread rates and flame lengths increase both with ambient oxygen concentration as well as free stream velocity. Quenching extinction has not yet been observed. The trends in spread rate and flame length agree with drop tower data. We have found, however that the magnitude of the heat flux as well as the size of the flames are considerably larger than observed. This discrepancy is presently being examined.

Future work includes examining the effect of gravity level on flame spread. Gas phase radiation is to be included. Finally, flame spread over thicker fuels will be examined. At some point, an unsteady model may be necessary to describe the flame spread process.

Publications

Ross, H.D., Sotos, R.G., and T'ien, J.S., "Observations of Candle Flames under Various Atmospheres in Microgravity," Comb. Sci. Tech. **75**, 155-160 (1991).

Sacksteder, K.R., "The Implications of Experimentally Controlled Gravitational Accelerations for Combustion Science," in *Proceedings of Twenty-Third Symposium (International) on Combustion*, The Combustion Institute, pp.1589-1596, (1990).

Grayson, G., Sacksteder, K.R., and T'ien, J.S., "An Experimental Study of Low-Speed Concurrent-Flow Flame Spread over a Thin Fuel," Central States Meeting, The Combustion Institute, April 22-24, 1991, Nashville Tennessee.

Ferkul, P.V. and T'ien, J.S., "Prediction of Flame Spread Over Thin Fuel in Low Gravity," Central States Meeting, The Combustion Institute, April 22-24, 1991, Nashville, Tennessee.

7. FUNDAMENTAL PHYSICS

Precise Viscosity Measurements Very Close to Critical Points

National Institute of Standards and Technology

Dr. Robert F. Berg

Dr. Michael R. Moldover

C-32001-K (NASA contact: Dr. R. Allen Wilkinson, LeRC)

October 1, 1990 - November 30, 1991

Objectives: Our objective is to accurately measure, in microgravity, the viscosity of xenon near its liquid-vapor point critical point. The space experiment will be the fourth in a series of studies of dynamic critical phenomena. These are: (1) theoretical studies, (2) viscosity measurements of binary liquid mixtures, (3) viscosity measurements of pure fluids in 1-g, and (4) viscosity measurements of xenon in low gravity.

Research Task Description: Near the critical temperature T_c the viscosity η is predicted to diverge as:

$$\eta \sim (T - T_c)^{-\gamma}.$$

Prior to this work, this functional form had been verified for binary mixtures near their consolute points but with apparent exponents γ ranging from the theoretical value (0.032) to as high as 0.042. In pure fluids near their liquid-vapor critical points, experimental values of γ were even more uncertain, in part because of gravitational stratification near T_c . The scientific task is to improve the measurement of γ , first on earth and then in a microgravity environment.

Progress to Date: We have developed a unique series of torsion oscillator viscometers devoted to critical point measurements with both low frequency and low shear operation in a thermostat with sub-millikelvin thermal perturbations. Their application to four binary mixtures narrowed the experimental range of the exponent to $0.040 < \gamma < 0.044$, a result inconsistent with theory. We have also measured the viscosity of CO_2 and xenon near their critical points. The xenon measurements will be an essential input for analyzing the results of the Zeno microgravity light scattering experiment. Analysis of the carbon dioxide and xenon data, which incorporated both modern crossover theory and 1-g stratification effects, gave exponents in the range $0.040 < \gamma < 0.042$. The consistency of our results for the binary mixtures and pure fluids is the most convincing evidence that both types of fluids are in the same dynamic universality class.

Measuring the viscosity in low gravity will allow the exponent γ to be determined without relying on the uncertain details of the theory for crossover from the noncritical to the critical region. However, the inherent vibration sensitivity of the existing 1-g viscometer requires an acceleration environment of less than $4 \times 10^{-8} \text{ g/Hz}^{1/2}$ near the viscometer's resonance at 1 Hz. In order to reduce the severity of this vibration specification, we have developed a prototype miniature viscometer consisting of a thin screen oscillating inside the sample container. At present, the nickel screen's 30 micron motion is driven by an applied oscillating magnetic field and detected capacitatively. By reducing the oscillator's mass by a factor of ten thousand, the instrument's sensitivity to low-level vibrations in orbit and to destructive vibrations during launch has been correspondingly reduced. This mass reduction was achieved without substantial changes in the shear rate, frequency of operation, or sheared area. Thus the new viscometer has approximately the same sensitivity to viscous forces as the older one.

Publications

R.F. Berg and M.R. Moldover, "Milliwatt Mixer for Small Fluid Samples," Rev. Sci. Instrum. **62**, 527 (1991).

Bubble and Droplet Phenomena in a Reduced Gravity Environment

The University of Toledo
Professor Kenneth J. De Witt
Douglas L. R. Oliver
Chain-Nan Yung
Jonathan L. Brockwell
Union Carbide Technical Center
NCC 3-34 (NASA Contact: Dr. An-Ti Chai, LeRC)
October, 1990 - October, 1991

Objectives: The objective of this research is to determine the merits/feasibility of a space experiment to study bubble and droplet phenomena. The distinguishing characteristic of the research is that it involves mass transport phenomena of a gas bubble dissolving in a liquid or of a liquid droplet evaporating into a gas. Multiple bubbles and movement due to thermocapillary or diffusocapillary migration are also considered. A numerical model has been developed which simulates the bubble dynamics under reduced gravity conditions and can be used to analyze bubble and droplet behavior. The results will yield information that will have application to many problems in physical chemistry and technology.

Research Task Description: The approach consists of experiments and numerical modeling. The modeling of the dissolution of a single bubble or of a moving bubble due to residual gravity effects has been accomplished. Present modeling involves incorporation of thermal and concentration gradients at the interface. Ground-based and drop-tower experiments include bubble and droplet motion due to an interfacial tension gradient and electrical effects. The justification for conducting the experiment in space is the elimination of large-scale buoyant bubble motion and free convective mass transfer effects that occur in normal gravity. These effects generally mask the molecular mass transfer and interfacial contributions to the dissolution process, and their elimination would allow a more fundamental understanding of the molecular mass transfer and interfacial efforts to be obtained.

Progress to Date: The Conceptual Design Review was held in January, 1990, and the Science Panel suggested broadening the scope of the experiment. The scope of the experiment has been since enlarged to include multiple bubbles and their interactions, along with movement due to thermal, concentration, and electrical effects. Numerical modeling of the unsteady motion occurring both inside and outside of a translating bubble or droplet has been completed.

Publications

Del Signore, D. M., Oliver, D. L. R., De Witt, K. J., and Rashidnia, N., "Experimental Demonstration of Diffusocapillary Flow in an Oil Droplet," presented at the AIChE/ASME/ANS/AIAA 27th National Heat Transfer Conference, Minneapolis, Minnesota, July, 1991. Published in AIChE Symposium Series 283, 127, 246-249, 1991. Also submitted to J. Colloid & Interface Science.

Yung, C. N., De Witt, K. J., Brockwell, J. L., and Chai, A. T., "The Transient Motion of a Spherical Fluid Droplet," accepted by Chemical Engineering Communications, 1991.

Determination of the Correlation Length in Helium II in a Microgravity Environment

University of Oregon
Dr. Russell J. Donnelly (P.I.)
Dr. Joseph J. Niemela
(NASA Contact: Dr. Donald Strayer, J.P.L.)

Objectives: The objective of this research is to measure finite size effects in the isobaric expansion coefficient near the lambda transition in liquid helium. Finite size effects are manifested as a rounding of the divergence in thermodynamic functions near a critical point as the correlation length increases toward the system size. We can thus test renormalization group theory predictions, universality assumptions, and boundary conditions.

We will measure the thermal expansion coefficient for liquid helium confined between parallel plates for a range of temperatures very near the lambda transition temperature (both above and below), a range of pressures from SVP to about 25 bar, and a range of plate separation distances. In order to measure finite size effects very near the lambda transition, or with a very well-characterized macroscopic geometry, we need a plate separation which would result in an extremely inhomogeneous sample in the earth's gravitational field; a microgravity environment eliminates this problem.

Research Task Description: We will measure the dielectric constant of helium confined between parallel plates as a function of temperature at constant pressure. Using the Clausius-Mossotti relation, we can calculate the density and thus the expansion coefficient of liquid helium. The experimental method involves two measurements, at a given temperature, of the balancing ratio of an audio-frequency ratio-transformer capacitance bridge, one with the sample capacitor empty and then filled with helium. Appropriate division of these ratios then yields directly the dielectric constant at that temperature.

The capacitor used to measure the dielectric constant is a parallel plate design operated as a three-terminal device in a 1 kHz ratio-transformer bridge. The spacing between the electrodes is determined by a precision shim which can be easily changed. An identical capacitor is also mounted on the experimental platform and is operated empty as a reference capacitor. We expect to vary the thickness of the shims between 5 microns and 50 microns.

We have the option of being able to make high-resolution measurements of the dielectric constant via microwave cavity resonance. The modifications to the apparatus involve installing quarter-wavelength coaxial heat sinks and constructing a new superconducting resonant microwave cavity (lead-plated copper) which can be attached to the experimental platform.

Initially, we will use only germanium thermometry for temperature control and measurement. This will allow us to easily cover a wide range of temperature and gain familiarity with the experiment. At this stage we will want to reconcile our results with older, published, data. High resolution measurements will be made after installing a paramagnetic salt thermometer identical to that used by John Lipa in his lambda-point heat capacity experiment, currently scheduled for flight in 1992.

Progress to Date: The new apparatus probe has recently been completed by our machine shop and we are now installing the low-resolution thermometry and leads and assembling the parallel plate capacitors. Most of the external support for the experiment has already been accomplished, including the construction of a crane for inserting and removing the cryostat, dewar, etc. from the experimental pit, a vibration isolation platform, supporting stand for the dewar, and installation of the major plumbing lines. We will soon begin a thorough leak-checking of the apparatus followed by an initial series of cool-downs designed to check the operation of the HeI refrigerator, the thermal stability and resolution of the various stages, and measurements of the capacitance of the empty parallel-plate capacitors (using a commercial

capacitance bridge). Any needed modifications to the apparatus which are indicated during these tests will be made prior to beginning measurements of the dielectric constant of helium.

Satellite Test of the Equivalence Principle (STEP)

W. W. Hansen Experimental Physics Laboratory, Stanford University
Professor C. W. Francis Everitt
Dr. Paul W. Worden, Jr.

Objectives: The objective of this research is to test the equivalence of inertial and passive gravitational mass in an Earth-orbiting satellite. We have done preliminary work and technology development with a ground-based version of the apparatus that could test equivalence to about one part in 10^{12} . Recent studies show that the satellite version of this experiment should have a sensitivity better than one part in 10^{17} of the total rate of fall. We envision the satellite experiment being performed as a joint NASA-ESA collaborative mission.

Research Task Description: The requirements for an orbital experiment have been well defined by studies using the Earth-based apparatus. The experiment consists in comparing the motions of concentric cylindrical test masses suspended in precision superconducting magnetic bearings and free to move along their common axis. The materials are chosen according to the best theoretical likelihood of an effect, and the masses, weighing about 1 kg each, are plated with superconductor to make them uniformly controllable. A position detector based on SQUID magnetometers measures both differential and common mode motion of the test masses. This makes an accelerometer sensitive to 10^{-14} cm/sec² or less. The remainder of the apparatus controls the positions of the test masses and isolates them from disturbances. Essential features include a drag-free spacecraft with acceleration levels of about 10^{-11} g/ $\sqrt{\text{Hz}}$; a mass positioning servo which uses the acceleration from the Earth's gravity gradient to center the masses on each other within 0.2 Å, a charge measurement and control system to limit the effects of cosmic ray charging; a method of controlling liquid helium sloshing; and superconducting shields for electromagnetic isolation.

We expect to be able to compare six test masses in a single six-month mission. The spacecraft would be an 800 kg free orbiter in a 550 to 750 km high, Sun-synchronous orbit. A version of this experiment performed on Shuttle would be severely limited by vibrations and drag; extreme care, together with some modification to the Shuttle attitude control system, could reach no better than 10^{-14} . The free orbiter should reach 10^{-17} and will be limited mostly by gas pressure effects and residual acceleration in the drag-free system.

Progress to Date: In November 1989, STEP was selected as a potential candidate for the European Space Agency's Next Medium-Sized (M2) Mission. Dr. Worden spent most of the summer and fall of 1990 participating in a feasibility study of STEP at the European Space Technology Engineering Center (ESTEC), Noordwijk, Holland, and he and Professor Everitt have formed the basis of a NASA-ESA collaboration on STEP by identifying and visiting interested groups in France, Germany, Italy, Switzerland and England. The ESTEC Study concluded that STEP is feasible. In April 1991, ESA selected four of the six candidate missions for a Phase A studies. The European Phase A study covers the STEP spacecraft and starts in January 1992. A parallel NASA Phase A study of the STEP payload will begin in October 1991. The studies continue for about a year and a half, and ESA will select the M2 mission in mid-1993. The present concept of the instrument, which uses the existing Lockheed "I.D." dewar as a baseline, was developed by Mr. Matthew Bye, the STEP Manager for Hardware Development. Mr. Bye has also developed the plans for the NASA Phase A study at Stanford.

Publications

Worden, P. W., "Satellite Test of the Equivalence Principle," in *Proceedings of First William Fairbank Conference*, 1991, in press.

Critical Transport Phenomena in Fluid Helium Under Low Gravity

Duke University

Horst Meyer

NAG 5-379 (NASA Contact: Stephen Castles, GSFC)

October 1990 - October 1991

Objective: These are to measure the shear viscosity η in ^3He - ^4He mixtures near the tricritical point ($T_t = 0.87\text{ K}$, $X_t = 0.67$) where X and T are the ^3He concentration and T the temperature. One could expect, by analogy with the behavior near the liquid vapor critical point, that η near (T_t, X_t) would also weakly diverge. In this case, the earth's gravity will produce a rounding of this divergence, because of the induced density gradients in the fluid layer. In the absence of gravity, this rounding should then disappear.

The first question to resolve is whether or not η near the tricritical point of helium mixtures really diverges. In 1973, Kawasaki and Gunton stated that η remains finite at the tricritical point, but did not elaborate. Kawasaki also predicted that for a multicomponent fluid near its tricritical point, the shear viscosity is "finite or at most weakly divergent". However measurements on a ternary mixture did clearly show a weak divergence of η , similar to that near the liquid-vapor critical point. For this purpose we have conducted a systematic study of the viscosity of ^3He - ^4He mixtures with particular emphasis on their superfluid transition curve and on the phase separation curve branches that join at the tricritical point.

Research Task Description: The viscosity is measured by means of a torsional oscillator operating in a continuous mode at the frequency of 150 Hz, as described in our recent publication [Wang *et al.*, J. Low Temp. Phys. **79**, 151 (1990)]. The horizontal fluid layer in our present cell is $\sim 0.04\text{ cm}$ high with a diameter of 5 cm and is contained in a thin walled beryllium-copper cell that oscillates around its axis with an extremely small amplitude detected electronically. The apparatus measures the product $(\rho\eta)$ where, in the *normal* phase of the mixture, ρ is the mass density determined in a small cell situated above the torsion oscillator. In the *superfluid* phase the relevant quantity is ρ/ρ_n , the density of the normal component. The ratio ρ/ρ_n is determined from different experiments, as quoted in our publication. Combination of $(\rho\eta)$, ρ/ρ_n and η measurements then finally gives the viscosity η .

Progress to Date: We have operated a new cryostat enabling us to extend the temperature range down to temperatures of 0.55 K to permit the investigation of the tricritical point. After an exhaustive test with dilute mixtures of ^3He in ^4He with $0.01 < X < 0.05$, we have carried out measurements of $(\rho\eta)$ for a number of mixtures near the tricritical point. For each series of experiments, a standard mixture was prepared, and once the measurements with each were completed, the concentration of the fluid was changed by small steps by means of removing a small fraction and adding a known amount of either ^3He or ^4He . In this way, we systematically investigated $(\rho\eta)$ for $0.30 < X < 0.80$ and for pure ^3He . We have completed these very time-consuming experiments and have analyzed most of them. A complete mapping of $\rho\eta$ and η in the T - X plane is underway.

The principal result is the evolution study of the singular behavior of η along the line of superfluid transition $T_\lambda(X)$. A small peak is observed for $X > 0.50$, but as the tricritical point is approached, this peak merges with the temperature-dependent background viscosity and can no longer be seen. Furthermore, there is no evidence of any singularity in η upon approaching the tricritical point along the two branches of the phase separation curve. This is unlike the behavior near the liquid-vapor critical point (CP). As a result, η not only is finite at T_t - in agreement with predictions - but also there is no evidence of a singular behavior.

Two papers have been written of which the first one has been submitted for publication and the second is scheduled for completion by December 1991. Oral presentations have been made at two meetings as listed below.

We have started a new project, the study of equilibration near CP of the vertical density profile in a fluid layer, subjected to the earth's gravity field. From previous experiments, the equilibration time appears to diverge as CP is approached, and a systematic study of this phenomenon has been proposed to NASA. Calculations for the design of a new cell in our cryostat have been made for an experimental study of ^3He and ^3He - ^4He mixtures.

The viscosity experiment has been carried out by two graduate students 1) Carl Howald, who completed his Ph.D. thesis and successfully passed his final examination in July, and who presently is working on the second viscosity paper and 2) Xi Qin, who is has been taking data, is analyzing them and is preparing the figures for the second paper. A new postdoctoral associate, Fang Zhong, has been put to work on the new equilibration experiment.

Publications

Meyer, H., Howald, C., Qin, X., and Nham, H.S., "Shear Viscosity of ^3He - ^4He Mixtures Near the Tricritical Point," in *Proceedings Low Temp. Physics Workshop, Eighth University of Oregon Conference on Low Temperature Physics*, Washington, DC, January 1991.

Howald, C., Qin, X., Nham, H.S., and Meyer, H., "Shear Viscosity Measurements of Liquid ^4He and ^3He - ^4He Mixtures near the Tricritical Point," *J. Low Temp. Phys.*, 1991 (submitted).

Presentations

Meyer, H., *Eighth University of Oregon Conference on Low Temperature Physics*, Washington, DC, January 1991.

Qin, X., Howald, C., and Meyer, H., *Bull. Am. Phys. Soc.* **36**, 1017 (1991).

8. EXPERIMENTAL TECHNOLOGY

Development of a Versatile Get Away Special Furnace for Microgravity Materials

GTE Laboratories Incorporated

Mr. Alfred Bellows

Mr. Glenn Duchene

Dr. David Matthiesen

Dr. Brian Ditchek

NAS3-25058 (NASA Contact: Dr. R. Lauver, LeRC)

October, 1990 - September, 1991

Objectives: The objective of this program is to design, fabricate and demonstrate a prototype growth system for an inexpensive, operationally simple capability for performing a variety of material processing experiments in space. The carrier chosen for the prototype design was a Get Away Special container. This carrier offers simple pre-flight procedures, low cost and, most importantly, the quick turn-around necessary to verify or refine an experiment.

Research Task Description: This program represents an extension and generalization of the development of the gradient freeze growth system, which, with sponsorship from NASA and the Air Force, is pioneering the utilization of GAS containers for the performance of serious materials science in space. Despite the enormous interest in materials processing in space there is, as of yet, no systematic procedure for assignment to individual Principal Investigators the associated equipment necessary for performing a variety of experiments in rapid sequence in the microgravity environment. A Bridgman-Stockbarger design with 13 cm hot and cold zones separated by a 2 cm gradient zone, was chosen as the prototype demonstration growth system. The furnace is designed for easy replacement of the molybdenum wound heater cores and thus, can be easily configured for other types of growth, such as, float zoning or isothermal studies. The growth system is capable of translating 14 cm with a wide range of growth speeds, selectable by hardware or software. The material chosen for demonstration was selenium doped gallium arsenide (Se/GaAs), which has a melting point of 1238°C. The fused quartz ampoule was held in a stationary molybdenum cartridge and the furnace was translated at rates of 2, 4 and 8 $\mu\text{m/s}$. The stationary position of the cartridge allowed access to both ends of the cartridge for *in-situ* diagnostic measurements. The Se/GaAs crystal was successfully grown and is currently being analyzed.

Progress to Date: A program task of a Conceptual Design Review, held at NASA-LeRC, was completed and the final report was delivered to NASA-LeRC.

Presentations

Bellows, A.H., Matthiesen, D.H., Duchene, G.A., and Chen, G., "A Versatile Get Away Special Furnace for Materials Processing in Space," to be presented at the AIAA 30th Aerospace Meeting in Reno, NV 1992.

Containerless Processing of Materials - Levitation Studies

Intersonics, Incorporated
Dr. Charles A. Rey
JPL Contract 958902

This report represents the completion of all FY'91 tasks performed by Intersonics Incorporated for NASA/JPL on Contract 958902. This work is part of NASA's continuing efforts in containerless processing of materials in space. It was divided into six different areas, which are briefly described below.

AGEMA NCTM System Brassboard

The development and construction of the AGEMA NCTM Brassboard was completed in FY'90. It was tested, evaluated, and calibrated in FY'91. This work was completed in December, 1990 and the final report was submitted in January.

An AGEMA Thermal Imaging System was evaluated to determine its feasibility for use as a temperature measurement device for the DPM Dual Zone Chamber, USML-1 Spacelab mission. Feasibility was established and a development project was carried out by Intersonics, Inc. to provide this capability. This development included the design and manufacture of custom objective optics for the AGEMA 870 scanner head, which was the sole component of the AGEMA system that was retained for this application. All support electronics were custom designed for integration compatibility with the DPM.

The NCTM System Brassboard was built, tested, and calibrated at Intersonics. It was integrated into a stand-alone system for testing purposes and is a fully operational system as such. Two electronic PWA's make up the support system and video digitizer for the AGEMA Thermal Imaging Scanner. A ground support simulating device provides access to the digital data to allow temperature measurement to take place prior to the conversion back to analog video. This device also allows the use of AGEMA support facilities by providing a regenerated analog video signal virtually unaltered by the digitization process.

Electromagnetic Processing Module (EPM)

The EPM was developed for testing aboard the KC-135 low gravity aircraft. It is based on the Stabilized Electromagnetic Levitator (SEL) technology being researched through a NASA-SBIR grant.

The SEL technology involves the use of multiple positioning coils and higher frequencies (typically 5 MHz to 20 MHz) than are used in other EM levitation techniques (typically < 1 MHz). The high frequencies provide very efficient heating of highly conductive samples but also allow positioning of poorly conducting materials. Multiple coils and the use of two or more frequencies simultaneously allow individual control of heating and positioning. The technique also provides the basis for closed loop specimen motion control for precise positioning and damping.

The following is a list of some key features of the device:

- o Independent control of specimen heating and positioning.
- o "O" ring sealed processing chamber suitable for evacuation and gas backfill.
- o Variable rate gas purge control for local sample purge during processing.
- o Sample cooling enhanced by gas quenching (variable).
- o Sample injection features
 - Semi-automated sample injection
 - Vacuum chuck sample holder
 - Samples contained in glass tube during processing

- Remote control inject and release function
- o Audio/Video system
 - Two orthogonal camera axes
 - Video multiplexor (up to 4 quadrant display)
 - Video cassette recorder
 - On screen data display (overlay)
 - Two video monitors, one in each rack
 - Two headsets for audio recording of test personnel
- o Optical Pyrometer
- o Data Acquisition System Acquiring
 - 4 pressure sensors
 - 3 accelerometers
 - 4 RF power signals
 - specimen temperature (optical pyrometer)
- o Large capacity bulk data storage unit
- o System designed to allow for expansion and upgrade of electronic and mechanical systems
- o Automatic frequency tuning of the EM levitation coil assemblies*

As this report is being prepared, the EPM is being prepared for the first set of KC-135 flights in December, 1991.

Remote Sample Deployment and Capturing

This project was to develop a design for an automated sample handling system that would be suitable for use in containerless processing applications. The primary design requirements included:

- o Compatibility with acoustic or electromagnetic levitation techniques,
- o Vacuum or gaseous operation
- o Suitability for space flight or sounding rocket applications
- o The ability to process a large number of samples

In addition to these requirements, a considerable amount of attention was paid to contamination control in the sample environment. Control of sample contamination was addressed for storage, insertion and retrieval. This was done from the standpoint of materials in direct contact with the sample, gaseous impurities, and cross contamination from other samples. To best satisfy these issues, separate, individually sealed specimen containers were designed. With this technique, a Principal Investigator can choose the gas and the materials which will contact the sample while it is in storage.

This system is adaptable for use on gaseous or vacuum processing chambers. It would provide a high degree of contamination control and provides a number of flexible design options for specific applications.

Sample Tracking and Beam Steering Brassboard

The objective of this task was to produce an electro-optical device for tracking a moving levitated sample, and for steering a laser beam to an optical target point on the sample. The purpose of this is to allow optical measurements to be carried out on a moving target by making it appear to be stationary. An additional requirement for this device was the ability to provide a continuous measurement of the target position in three dimensional space.

The completed brassboard was developed based on a number of design requirements and operating parameters. They are as follows:

- o The device must:
 - Locate an optical target point (specular reflection from a spherical sample) and track that point as the sample moves.
 - Introduce a generic laser beam to the target point and steer the beam as the target moves.
 - Provide an output signal proportional to the instantaneous target position for the three axes of motion.
- o The device must operate independently of:
 - Levitation system
 - Target or furnace wall temperature
 - Wavelength of interrogating laser beam from .6 to 2 μm .
- o Sample Characteristics:
 - Nominally spherical
 - Specular or partially specular surface
 - Opaque or semi-opaque at tracking laser wavelength
 - Up to 10 mm diameter
- o Sample Motion: $\sim \pm 10$ mm

For use with a device such as DAPP, it can operate in either a steering or tracking mode. However, as a beam steering device, the benefits of using this device with DAPP can be realized.

Advanced Video System

A feasibility study was undertaken to develop a conceptual design for a video system with the capability of replacing currently used cine film systems such as the system being used in the DPM. The video system design goal was to obtain the same 400 fps frame rate and the same resolution that is achievable with the 16 mm camera using Kodak 7292 color film. The application was to provide real time image reproduction of space lab or space study experiments via data downlink but to still retain the high frame rate and resolution characteristics of the cine film system. In addition, substantial bulk and weight savings may be realized if film storage was not required and crew maintenance time could be reduced.

The directed approach for development of this system was to investigate the feasibility of utilizing only commercially available components. The system consists of a video camera, an analog to digital converter, a short term memory buffer, and long term bulk data storage.

Division of Amplitude Polarimetric Pyrometer (DAPP)

This task was part of the ongoing project to develop the DAPP technology. Its objective was to develop an advanced instrument, operating at the HeNe laser wavelength (.633 μm), which would incorporate pyrometric measurements and design improvements based on previous DAPP work. The delivered device provides the following features and upgrades:

- o Optical pyrometer and polarimeter for measurement of radiance temperature and sample emissivity.

- o Position detector for alignment purposes.
- o Video camera for observation of the specimen and alignment of the pyrometer.
- o Acousto-optic modulator to replace chopper wheel for AC laser light operation.
- o Synchronous detection of modulated laser beam for improved signal to noise ratio
- o Leveling and monitoring capability
- o Automatic shuttering capability

In addition to these features, the optical layout has been redesigned and some components replaced. The same is true for the electronic system and modifications have also been made to the software. The result is a more reliable and user friendly device.

Electrostatic Containerless Processing Technology

Jet Propulsion Laboratory
Dr. Won-Kyu Rhim
In-House

Objectives: There are several important characteristics of the electrostatic positioning method when viewed against other positioning methods such as the acoustic and the electromagnetic methods. The developmental effort of electrostatic positioning system has been justified on the following basis; (i) quiescent sample positioning is provided by three dimensional feedback control and it does not induce internal flow in the melts, vibration, oscillation, or rotation; (2) it operates in 1-g as well as in reduced-g environments; (3) both conducting and nonconducting sample materials can be levitated; (4) it operated both in vacuum and controlled gas environments; (5) sample positioning and heating are completely decoupled; and (6) wide open structure allows clear sample viewing. Since the first successful demonstration of electrostatic levitation in 1983, the system has been improved and modified in anticipation of various experiments. Initially, we have developed electrostatic positioners which are operable in ambient condition. Feedback-controlled *tetrahedral levitator* and *electrostatic acoustic hybrid system* have opened up the possibilities for containerless crystal growth from solutions as well as the various charged drop-dynamics experiments in micro-gravity environment.

Progress to Date: Development of high temperature electrostatic positioning technology in vacuum has been progressing very well. High temperature sample charging methods have been extensively investigated and various charging methods have been identified. Using photoelectric and thermionic charging methods various metal samples could be levitated and heated to high temperature. In a test experiment, we succeeded in melting and solidification of aluminum and tin samples during levitation. In the case of a titanium sample, we observed stable levitation as the sample was heated beyond 1200°C. As it stands now, this technology looks very promising for the containerless processing of metals and alloys in 1-g and reduced-g environments.

The main thrust during FY'92 will be to continue developing the high temperature electrostatic positioning system for the microgravity application. The main activities will include: (a) melting and solidification of Co, Ge, Fe, and Ni; (b) completion of KC-135 tests of the High Temperature/High Vacuum Electrostatic Positioner; (c) development of an ellipsoidal mirror furnace to achieve high temperature and more uniform sample heating; (d) determination of surface tension of undercooled melts through sample rotation; and (e) development of an adaptive feedback control software.

Publications

Rhim, W-K., Chung, S.K., and Elleman, D.D., "Electrostatic Sample Positioning and Drop Dynamics," in *Proceedings of 7th European Symposium on Materials and Fluid Science in Microgravity*, ESA-295, 1990, pp. 629-637.

Sridharan, G., Chung, S.K., Elleman, D.D., and Rhim, W-K., "Optical Sample-Position Sensing for Electrostatic Levitation," in *Proceedings of SPIE Space Material Processing Conference* 1118, 160 (1989).

Rhim, W-K., and Chung, S.K., "Containerless Protein Crystal Growth Method," *J. Cryst. Growth* 110, 293 (1991).

Rhim, W-K. and Chung, S.K., "Isolation of Crystallizing Droplets by Electrostatic Levitation," in *METHODS: A Companion to Methods in Enzymology* 1, 118-127 (1990).

Colvin, M., Chung, S.K., Hyson, M.T., Chang, M., and Rhim, W-K., "A New Method for the Production of Large Spherical Monosized Polymer Particles for Biomedical and Chromatographic Applications," J. Poly. Sci. A: Poly. Chem. **28**, 2085-2095 (1990).

Rhim, W-K., Hyson, M.T., Chung, S.K., Colvin, M.S., and Chang, M., "Monodisperse, Polymeric Microspheres Produced by Irradiation of Slowly Thawing Frozen Drops," U.S. Patent 4,981,625, January 1991.

B. FLIGHT EXPERIMENTS

1. ELECTRONIC MATERIALS

A Comparative Study of the Influence of Convection on GaAs

GTE Laboratories Incorporated

Dr. Brian Ditchek

Dr. David Matthiesen

Mr. Alfred Bellows

Mr. Glenn Duchene

NAS3-24644 (NASA Contact: Dr. R. Lauver, LeRC)

October, 1990 - September, 1991

Objectives: The objective of this study is to determine the effects of buoyancy driven fluid flow on the properties of melt grown GaAs crystals. This program is sponsored by NASA, GTE Laboratories and the United States Air Force.

Research Task Description: A highly efficient gradient freeze growth system has been developed to grow a one inch diameter by 3.5 inch long selenium doped gallium arsenide crystal (Se/GaAs). Growth systems, with and without, interface demarcation by current pulsing (IDCP) have been designed, fabricated and tested. Identical ground based versions of the furnaces and control systems have been fabricated and tested. These ground based systems have been used for extensive comparative studies. The two flight qualified growth systems have been incorporated into a fully self-contained payload which was launched as part of the Get Away Special program.

Progress to Date: The ground based crystal growth runs in the bottom-seeded vertical Bridgman, with and without IDCP, as well as the horizontal Bridgman growth, with and without IDCP, have been completed. The ground based crystal growth runs in the bottom-seeded vertical Bridgman, with and without IDCP, in the presence of a 20kG axial magnetic field have also been completed.

The payload was launched as part of the GAS Bridge on STS-40 (Columbia) on June 5, 1991. The first crystal growth run was a complete success, with the expected 5.7 cm of growth in space. Temperature, power and accelerometer data were recorded and are currently being analysed. A preliminary report has been delivered to NASA-LeRC.

Presentations

Matthiesen, D.H. and Bellows, A.H., "Microgravity Acceleration Measurements Aboard the Get Away Special Bridge During STS-40," to be presented at the AIAA 30th Aerospace Sciences Meeting, Reno, NV 1992.

Compound Semiconductor Growth in Space

NASA Langley Research Center
Dr. A. L. Fripp
Mr. W. J. Debnam
Dr. I. O. Clark
Dr. R. K. Crouch, NASA Headquarters

Objectives: The objectives of this research are to determine the effects of gravity driven convection on the growth parameters and crystal properties of compound semiconductors.

Research Task Description: The Microgravity Science effort at the Langley Research Center is centered on the growth of the compound semiconductor alloy, lead tin telluride. Lead tin telluride is a substitutional alloy of lead telluride and tin telluride that is miscible over the entire compositional range. The semiconductor properties of this material are dependent on the ratio of the two components and consequently, the uniformity of an array of devices is dependent on good compositional control.

This material was chosen for microgravity research for a number of reasons. Lead tin telluride is not only a useful semiconductor material which has been used for construction of infrared detectors and tunable diode lasers but it also has a similar phase diagram to other compound semiconductors of interest such as mercury cadmium telluride and mercury zinc telluride.

Lead tin telluride is very interesting from a purely scientific point of view in that it is both solutally and thermally unstable, but in a one dimensional analysis with growth axis parallel to the gravity vector, only one instability works, per orientation, at a time. This double convective instability cannot be made stable by balancing thermal and solutal expansion in a high temperature gradient. Lead tin telluride is amenable to study for it is easily compounded; it has a relatively low vapor pressure; it is single phase and there is existing, though limited, literature on its growth and properties. The desired growth mode is, of course, one in which convection is zero so compositional steady state can be reached. However fluid dynamic calculations have shown that finite convection exists in the physical configuration used in crystal growth experiments even at 1×10^{-8} Earth gravity if there is a density gradient orthogonal to the gravity vector. However, due to the residual atmosphere the minimum gravity level expected on the Space Shuttle is roughly the order of 1×10^{-7} Earth gravity. Hence experiments are designed such that interface movement ie, growth rate, is greater than the anticipated fluid velocity.

Other work at Langley ranges from fundamental studies involving the direct measurement of both steady and oscillatory fluid flow in a crystal growth melt to efforts in furnace design and calibration. Also of interest is an effort to measure the melt-solid interface shape and position as this is of great importance to understanding the crystal growth process.

Publications

A.L. Fripp, W.J. Debnam, R.K. Crouch, R.T., Simchick, S.K. Sorokach, W. Rosch, D.J. Knuteson and P.G. Barber "Ground Based Preparation for Microgravity Growth of Alloy Semiconductors," AIAA Aerospace Sciences Meeting, Reno, Nevada; January 7-10, 1991, Paper AIAA-91-0511.

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- B. Sears, R. Narayanan, T.J. Anderson and A.L. Fripp, "Development of a Flow Visualization Technique for Characterizing Convection in Liquid Metals- Part I, Measurement of Oxygen Diffusivity in Tin," Met. Trans., 1991 (submitted).
- B. Sears, R. Narayanan, T.J. Anderson and A.L. Fripp, "Convection of Tin in a Bridgman System- Flow Characterization by Effective Diffusivity Measurements- Part II," J. Cryst. Growth, 1991 (submitted).
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- A. Nadarajah and R. Narayanan, "Comparison Between Morphological and Rayleigh-Marangoni Instabilities," in *Dissipative Structures in Transport Processes and Combustion*, Volume 38 (D. Meinkohn, ed.), Springer- Verlag, 1990, p. 215.
- C. Wagner, R. Friedrich and R. Narayanan, "Gradational Effects and Some Extended Results for Bilayer Convection of the Rayleigh Marangoni Type," J. Phys. Fluids A, 1991 (submitted).

Presentations

- W. Rosch, A. Fripp, W. Debnam, R. Simchick and S. Sorokach, "Damage of Fine Diameter Platinum Sheathed Type R Thermocouples at Temperatures Between 950 and 1100°C," *7th International Symposium on Temperature, Its Measurement and Control in Science and Industry*, April 28- May 1, 1992, Toronto, Ontario, Canada
- S. Sorokach, W. Rosch, A. Fripp and W. Debnam, "Errors in Small Diameter Sheathed Type K and N Thermocouples in High Temperatures Gradients," *7th International Symposium on Temperature, Its Measurement and Control in Science and Industry*, April 28- May 1, 1992, Toronto, Ontario, Canada

Solution Crystal Growth in Low-g

Alabama A & M University
Dr. R.B. Lal
Dr. A.K. Batra
Dr. W.R. Wilcox, Clarkson University
Dr. J.D. Trolinger, MetroLaser
NAS8-36634 (NASA Contact: Mr. Rudolph Ruff, MSFC)
September 25, 1986 - December 31, 1992

Objectives: The objectives of this research project are: 1) to grow crystals of triglycine sulfate (TGS) using modified Fluid Experiment Systems (FES); 2) to perform holographic interferometry tomography of the fluid field in three dimensions; 3) to study the fluid motion due to g-jitter by multiple exposure holography of tracer particles, and 4) to study the influence of g-jitter on the crystal quality.

Research Task Description: This project involves a reflight of an experiment, "Solution Growth of Crystals in Zero-g," flown earlier on Spacelab-3 mission. Two experiment runs are now planned for the First International Microgravity Laboratory (IML-1) to be flown in January 1992. The first experiment run is primarily aimed towards the study of the fluid motion using multiple exposure holography of tracer particles. This run is expected to last about 20 hr. The second run which will be about 40 hr will be used to grow a TGS crystal using a predetermined polynomial for the temperature programming of the seed crystal. The orientation of seed crystals for these runs will be either (001) or (010). Experiments have been completed to determine the proper shape and size of the seed crystals for the flight. The flight seed crystals have been mounted and the cells have been filled with TGS solution and Shipped to Kennedy Space Center.

Experiments are conducted in the Ground Control Experiments Laboratory (GCEL) where a test cell similar to the flight is being used to grow crystals using a laboratory version of the optical system. Experiments have been conducted to determine the proper size, type, and number density of particles that should be used in the FES to monitor fluid flows. During GCEL runs, holograms are recorded with particles of 300 μ , 400 μ , and 600 μ sizes. Different size particles are expected to diffuse at different rates enabling us to identify the particles. Proper design of holographic optical elements (HOE) has been completed. The flight windows of the FES test cells have been coated with proper HOE's. The incorporation of the HOE's will provide three independent views of the fluid field and the growing crystal. The central part of the optical windows is left uncoated for real-time monitoring using schlieren technique.

The space grown crystals will be characterized for defects by high resolution synchrotron radiation imaging and laser scattering techniques. To evaluate the device performance of the crystals, various properties, such as, dielectric constant and loss, spontaneous polarization, hysteresis loop, and pyroelectric coefficient will be measured. Infrared detectors will be fabricated and tested. All these parameters will be compared with the seed crystals and other crystals grown on ground in our laboratory and elsewhere.

Publications

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Orbital Processing of High-Quality CdZnTe Compound Semiconductors

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August 3, 1990 - May 31, 1993

Objectives: The objectives of this research are: (1) to quantitatively evaluate the influences of gravitationally-dependent phenomena (convection and hydrostatic pressure) on the chemical homogeneity and defect density of CdZnTe crystals grown by the Seeded Bridgman-Stockbarger technique; (2) to develop high-fidelity process models of the Seeded Bridgman-Stockbarger crystal growth process, including heat transfer, fluid flow and solute redistribution, and thermo-mechanical stress; (3) to develop, conduct, and evaluate a critical flight experiment in the CGF on USML-1, including a quantitative comparison of the flight sample with 1-g simulation and ground-best samples.

Research Task Description: This experiment is to advance the understanding of Seeded Bridgman-Stockbarger crystal growth and the influences of hydrostatic, wetting, and gravitationally-dependent thermo-solutal convection on the structural and chemical quality of alloyed compound semiconductors. In the course of this program we will advance the state of seeding technology and seriously attempt to use process models for process optimization, improving the chemical homogeneity and defect densities of the material and predetermining boule orientation, increasing primary yield of high quality material for infrared applications.

Progress to Date: Three seeded CdZnTe development test experiments have been conducted in ground based facilities at Grumman and 3 seeded test experiments and 1 flight simulation experiment have been conducted in the Crystal Growth Furnace (CGF) at the Ground Control Experiments Laboratory in Huntsville. Ampoules have been fabricated for 2 additional flight simulation experiments, 1 vibrational test, 6 thermal safety tests, and 1 flight experiment and 2 flight spares.

Very high quality, untwinned, seed crystals have been machined from CdZnTe grown at Grumman. These seeds, and the parent boules, have been thoroughly characterized using: optical and infrared microscopy, differential etching techniques, x-ray Laue analysis; x-ray four crystal rocking curve analysis, x-ray precision lattice parameter analysis, synchrotron x-ray white beam and monochromated topography, fourier transform infrared spectroscopy, and optical reflectance spectroscopy. We will use these seeds throughout the program. The GCEL experiments have served to empiricize the CGF thermal model and to develop seeding technology. We believe that both of these technologies are mature at this point. The flight simulation experiments will serve as a direct one-g comparative baseline for the flight experiment sample.

Two seeded Bridgman-Stockbarger experiments on the Grumman programmable multizone furnace have been conducted, and will continue throughout the year. The second one-g simulation experiment on the GCEL is scheduled to occur in February 1992. The flight ampoules will be delivered for the USML-1 flight in May 1992. The flight is expected to be conducted in May/June 1992 in STS-50.

The time between flight sample delivery and the return of the flight experiment will be spent developing and empiricizing the thermal, fluid-flow/solute redistribution, and thermomechanical stress

models of the Seeded Bridgman-Stockbarger crystal growth process and producing an optimized one-g CdZnTe crystal for comparison with the CGF micro-g flight and 1-g simulation samples.

Two crystals of V-doped CdTe have been grown at Rockwell Science Center, by co-investigators, and these are presently being machined and polished for opto-electronic analysis. These samples are representative of the material that we intend to evaluate on our reflight opportunity. Rockwell Science Center personnel, in collaboration with Clarkson University co-investigators, have also conducted short duration micro-g wetting studies on KC-135 aircraft.

Clarkson University co-investigators have conducted Ge crystal growth studies in support of the thermal model of seeded Bridgman-Stockbarger crystal growth. The model for seeded growth is expected to be completed in 1992 and will contribute to the flight sample analysis.

University of Alabama in Huntsville personnel have completed an analysis of the experiment flight orientation sensitivity. They will also complete a predictive analysis of the expected flight conditions and the resulting solute redistribution (radial and longitudinal chemical macrosegregation) within the flight sample. Obviously, this will contribute to the flight sample analysis, and will be compared to the one-g results. We anticipate that the sample will be of unprecedented uniformity.

Lastly, the thermo-mechanical stress within the growing and cooling CdZnTe crystal has been modelled at Grumman. The principal influences on defect density have been shown to result from interface shape, thermal gradients, heat extraction, and cooling rate. The anticipated dewetting during the flight experiment has been shown to be beneficial. However, x-ray synchrotron strain contour mapping of the CGF development test samples has shown that the cooling rates during the flight experiment are not ideal, and that a higher than optimum strain level will be typical of the flight sample. This results from flight time-line restrictions on the shared CGF flight facility.

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Growth of Solid Solution Crystals

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In-House

Objectives: The major objective of this research is to establish the limitations imposed by gravity during growth on the quality of bulk solid solution semiconducting crystals. An important goal is to explore the possible advantages of growth in the absence of gravity. The alloy system being investigated is $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ with x -values appropriate for infrared detector applications in the 8 to 14 μm wavelength region. Both melt and Te-solvent growth, as well as growth in a magnetic field, are being considered. The study consists of an extensive ground-based experimental and theoretical research effort required to define the optimum experimental parameters of the planned flight experiments. $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ is representative of several II-VI alloys which have electrical and optical properties that can be compositionally tuned to meet a wide range of technological applications to optical computing and communications as well as the national defense.

Progress to Date: A series of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ alloy ingots ($0 \leq x \leq 0.6$) has been grown from pseudobinary melts by a vertical Bridgman-Stockbarger method using a wide range of growth rates and thermal conditions. Precision measurements were performed on the ingots to establish compositional distributions for the ingots. Growth rates and thermal conditions required to obtain the desired growth interface shape have been established for the system.

To assist the interpretation of the results and the selection of optimum in-flight growth parameters, the pseudobinary phase diagram ($0 \leq x \leq 1$), liquid and thermal diffusivities ($0 \leq x \leq 0.3$), and the specific columns as a function of temperature ($0 \leq x \leq 0.15$) have been measured. From these measurements and other available data, the heat capacity, enthalpy of mixing, and the thermal conductivity of pseudobinary melts have been calculated using a regular associates solution model for the liquid phase. A one-dimensional diffusion model that treats the variation of the interface temperature, interface segregation coefficient, and growth velocity has been used to establish effective diffusion constants for the alloy system. Theoretical models have been developed for the temperature distribution and the axial and radial compositional redistribution during directional solidification of the alloys. These models are sufficiently accurate that they will be used along with the experimental results to select parameters for the first flight experiment to be flown on the USMP-2 mission using the Advanced Automatic Directional Solidification Furnace (AADSf).

A microscopic model for the calculation of charge-carrier concentration, Fermi energy, and conduction-electron mobility as functions of x , temperature, and both ionized and neutral defect densities has been developed. For selected samples, measurements were performed of electron concentration and mobility from 10-300K. The experimental data were in reasonably good agreement with theory and were successfully analyzed to obtain donor and acceptor concentrations for various processing conditions.

The crystal growth studies have been extended to include additional Hg-based II-VI alloys. Large crystal ingots of HgZnTe and HgZnSe have been successfully grown by the Bridgman-Stockbarger method and a detailed theoretical analysis of the measured axial compositional distribution in the ingots was used to establish for the first time effective HgTe-ZnTe and HgSe-ZnSe interdiffusion coefficients for the molten alloys. Both the Te and Se-based alloys showed improvements in lattice strength resulting from the substitution of Zn into the respective Cd lattice sites. In particular, measured electrical and optical properties, thus providing the first direct experimental confirmation for predicted improvement in lattice stability against point-defect formation resulting from Zn-additions.

Microhardness measurements were performed on selected wafers cut from the various alloy crystals. For each alloy, measurements were made for several alloy compositions. The microhardness results support the above conclusions. Sample cartridges needed for the integration of the samples into the space flight hardware are being developed. $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ between 2 and 5 kg. Precise compositional mapping of the ingots showed that the fields had significant effect on the composition for the entire field range.

The development and fabrication of the AADSF flight unit and a Ground Control Experiment Laboratory unit have been completed and are being readied for flight qualification testing. Gallium doped germanium samples have been grown in the AADSF prototype furnace. Mechanical pulsing was used to delineate the interface, and several different temperature zone settings were used. Concave, flat and convex interface shapes were realized and compared to thermal models of the furnace.

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Crystal Growth of Selected II-VI Semiconducting Alloys by Directional Solidification

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Objectives: The three-fold objectives of the investigation are: 1) to determine the relative contributions of gravitationally-driven fluid flows to the compositional redistribution observed during the unidirectional crystal growth of selected solid solution semiconducting alloys having large separation between the liquidus and solidus of the constitutional phase diagram, 2) to ascertain the potential role of irregular fluid flows and hydrostatic pressure effects in generation of extended crystal defects and second-phase inclusions in the crystals, and 3) to obtain a limited amount of "high-quality" materials needed for bulk crystal property characterizations and for the fabrication of various device structures needed to establish ultimate material performance limits. The effort includes both Bridgman-Stockbarger and solvent growth methods, as well as growth in a magnetic field. The work emphasizes $\text{Hg}_{1-x}\text{Zn}_x\text{Te}$ and $\text{Hg}_{1-x}\text{Zn}_x\text{Se}$ semiconducting alloys because of their potential applications for infrared detection and imaging in the 5 to 30 wavelength region. The initial experiments will use the Crystal Growth Furnace (CGF) to be flown on the first U.S. Microgravity Laboratory (USML) mission. The investigation complements the work being done on crystal growth of the $\text{Hg}_{1-x}\text{Zn}_x\text{Te}$ alloys using the Advanced Automatic Directional Solidification Furnace crystal growth system.

Progress to Date: A number of $\text{Hg}_{1-x}\text{Zn}_{1-x}\text{Te}$ ($x=0.15$) ingots have been grown by a vertical Bridgman-Stockbarger method using existing laboratory crystal growth hardware. Precision measurements were performed to establish detailed compositional distribution maps for the crystals. Fitting these and previously obtained data to numerical solutions to a one-dimensional diffusion model was used to establish estimates for the effective liquid-liquid diffusion constants for both the HgZnTe and HgZnSe alloys. Rapid quenching experiments were performed to establish correlations between thermal conditions and melt/solid interface shapes and to obtain values for the interface segregation coefficient for the $\text{Hg}_{0.15}\text{Zn}_{0.15}\text{Te}$ alloy. Three samples of the alloy have been also processed in the Ground Control Experiment Laboratory unit of the CGF. The results from these experiments have been used to fine tune optimum temperature settings for the flight experiment. The measured compositional profiles for the samples indicated that back-melting of the samples to the desired locations, a critical requirement for the flight experiment, have been successfully accomplished. Several flight samples have been prepared and delivered for flight integration and the results have been published.

Detailed thermal modeling of the growth process, measurements and modeling of the electrical and optical properties, and determination of some of the pertinent thermophysical and thermodynamic properties for the HgZnTe alloy system are all in progress. Preliminary growth experiments in the presence of magnetic fields have also been performed.

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The Study of Dopant Segregation Behavior during Growth of GaAs in Microgravity

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NAS8-38148

October, 1990 - September, 1991

Objectives: The objectives of this program are to investigate gravitational and thermal techniques for obtaining complete axial and radial dopant uniformity of the selenium dopant during crystal growth of gallium arsenide (GaAs). These techniques include controlling the thermal conditions to obtain a flat interface shape and a steady state growth rate and, most importantly, growth in the microgravity environment afforded by the Crystal Growth Furnace (CGF) in the first United States Microgravity Laboratory (USML-1).

Research Task Description: Crystals of GaAs, 16.5 cm long by 1.5 cm diameter, will be supplied by GTE Laboratories to NASA for growth in CGF on USML-1. These crystals will be doped with selenium to approximately $1 \times 10^{17}/\text{cm}^3$. As supplied to NASA, these crystals will be hermetically sealed in a specially designed fused quartz ampoule. This ampoule will then be sealed into an experiment cartridge, which on orbit will be loaded into the furnace system for growth. The large hot zone length of the CGF (20 cm) should allow, for the first time in microgravity, the achievement of steady state growth rates. After flight, extensive characterization of the electrical and structural properties are planned.

Progress to Date: Three ampoules containing GaAs have been tested at GTE Laboratories. In addition, two alumina thermal probes and three GaAs samples have been tested in the Ground Control Experiment Laboratory (GCEL). The GCEL contains a ground based equivalent of the flight unit, which has been delivered to the Kennedy Space Center for integration into the United State Microgravity Laboratory.

Publications

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Double Diffusive Convection During Growth of Lead Bromide Crystals in Space

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Objectives: The main objectives of this program are to grow lead bromide crystals doped with silver bromide under diffusion controlled conditions, and to understand the origin of thermal and solutal convection during vertical crystal growth. Lead bromide holds great promise in application for acousto-optic devices, for optical signal processing, and for optical spectrum analyzing systems. To achieve low optical and acoustic attenuation and to maximize device efficiency, a high degree of uniformity of refractive index is crucial. Lead bromide is doped with silver bromide to reduce the cracking because of the destructive phase transformation during cool down process. In earth grown crystals thermal and solutal convection during crystal growth can degrade crystal chemical and optical homogeneity significantly. This program is aimed to (a) test this hypothesis in normal and reduced gravity and to relate the results to the growth of this device grade crystals, (b) demonstrate lead bromide acousto-optic crystals with unparalleled optical perfection for advanced device application and (c) provide basic data on convective behavior in crystals grown by the commercially important Bridgman process.

Research Task Description: Two sets of tasks were required to achieve the goals of ground-based experiments. The effect of thermal convection on crystal quality was evaluated by growing several crystals at different thermal convection levels. The convection level was changed by changing the aspect ratio. The crystal quality was evaluated by measuring the optical distortion, birefringency and light scattering. The solutal convection level was changed by adding different amounts of silver bromide in the lead bromide at fixed aspect ratio.

Base-line experiments for space equipment were carried out in a two zone furnace with sealed ampoules. Experiments on crystal growth have begun in the CVTE furnace with ampoules identical to that of space ampoules. To avoid variations due to impurity content and aspect ratio, large batch of material and ampoules with identical sizes are required. The parallel crystal growth work in the CVTE and Westinghouse glass furnaces will provide ground based data needed to compare with crystals grown in space.

Progress to Date: The lead bromide crystals are very anisotropic and the growth rate varies with crystal orientation. We grew all the crystals in the [010] direction, the orientation needed by physicists for device fabrication. As mentioned in the previous section, thermal Rayleigh and solutal Rayleigh numbers were varied by changing the ampoule aspect ratio and dopant concentration. The temperature, speed and cooling conditions were identical in all experiments. These boundary conditions imply that observed changes in the quality should be due to thermal and solutal convection. With the increasing thermal convection level we observed high optical distortion, large number of optical fringes and high light scattering. We could change the thermal Rayleigh number approximately by two orders of magnitude. Similarly, with increasing solutal convection the optical crystal quality deteriorated.

At this stage four growth ampoules identical to that of space ampoules have been prepared. We are growing two crystals in the CVTE furnace and two in a Westinghouse glass furnace. In addition, we are preparing ampoules for the flight experiment and two more experiments in the CVTE.

Publications

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Vapor Crystal Growth of Mercuric Iodide

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Objectives: Single crystals of mercuric iodide are used in high-efficiency x-ray and gamma ray detectors operating at ambient temperature. Optimal operation of the devices is determined to a large degree by the density of structural defects in the single crystalline material. Since there are strong indications that the quality of the materials is degraded by the effects of gravity during the growth process, a program was initiated to grow one or more crystals of mercuric iodide in the reduced gravity environment of space.

Specifically, there are two reasons to perform the space experiments:

1. Single crystals of mercuric iodide are prone to slippage under the effect of gravity, especially at the elevated growth temperatures, with a concurrent decrease in structural quality.
2. It is not clear what effects convection flows in the vapor phase have on the growth rate and the homogeneity of the crystals. Growth in reduced gravity would provide information regarding these questions.

Research Task Description: The first experiment, performed during the flight of Spacelab 3 (1985), was highly successful in the sense that all scientific objectives were fulfilled. The structure of the space-grown crystal was more homogeneous and the critical electronic properties were increased by a factor of seven compared with the best earth-grown crystals.

Progress to Date: The preparations to perform the experiment on the IML-1 flight are complete. On the basis of ground-based experiments and the experience gained during the Spacelab 3 flight it will be possible to use optimal parameters for crystal growth. It is expected that in the short flight time available a crystal can be grown large enough so that a sufficient amount of material is available for extensive analysis.

Vapor Growth of Alloy-Type Semiconductor Crystals

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NAS8-38143
October 1990 - October 1991

Objectives: The present effort is an important part of a continuing research program directed towards the investigation of basic vapor transport phenomena and of crystal growth properties of electronic materials. The primary objectives of ground-based studies are the development and definition of optimum experimental parameters for flight experiments. The specific experiments to be performed in a microgravity environment include the investigation of vapor transport and crystal growth phenomena of the $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ - HgI_2 system. Emphasis for this system is on the mass flux, on the seeded growth of bulk crystals, and on the growth of epitaxial layers on CdTe substrates. The above experiments are performed in closed, fused silica ampoules.

The objectives of the $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ experiments are to determine the positive effects of microgravity on vapor phase crystal growth of ternary, alloy-type materials in terms of chemical and structural microhomogeneity, and of electrical properties of crystals grown in space. In order to achieve these goals it is necessary to perform corresponding crystal growth studies on ground which will serve as a comparative basis for the meaningful analysis of microgravity effects. Thus, the objectives of ground-based studies are to determine the effects of gravity-driven convection on mass transport rates and on crystal morphology for the bulk and epitaxial growth of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ crystals. These objectives include the thermodynamic and fluid dynamic analysis of the mass transport processes, and the characterization of grown crystals in terms of defects and of electrical properties. The ultimate goal is a quantitative correlation between crystal morphology, electrical properties, and growth parameters for this system.

Research Task Description: Earlier studies demonstrated the effects of gravity-driven convection on mass flux and morphology of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ bulk crystals. The continued experimental tasks are directed towards the investigation of the effects of temperature profile and of the geometry of the condensation region of the ampoule on mass flux and crystal morphology. In addition, the effects of aspect ratio of the ampoule on the mass transport and crystal growth properties will be investigated. The ground-based tasks include the investigation of the above effects for different orientations of the density gradient with respect to the gravity vector, and for different transport agent (HgI_2) pressures and temperature profiles. An important aspect of this effort is the development of a combined CVT-seeding method for the bulk growth of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$. The crystal characterization techniques include various microscopic and x-ray diffraction methods, chemical etching, and electrical measurements. The ground-based studies are designed to optimize experimental parameters for flight experiments.

The major tasks of ground-based studies of the epitaxial growth of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ layers by chemical vapor transport reactions involve systematic investigations of the growth rate, morphology, homogeneity, and electrical properties of HgCdTe layers. These studies include measurements of the effects of substrate orientation, of temperature profile effects, and of transport agent pressure on the above properties. They are performed under horizontal and vertical stabilizing conditions with the goal to observe the effects of convective interferences on layer morphology and properties. The results of ongoing ground-based studies are continuously evaluated and are used for the systematic improvement of growth parameters with the important goal to define optimum experimental conditions for the microgravity experiments of this system.

In addition to the experimental tasks, theoretical efforts involve the quantitative thermodynamic analysis of the system under investigation, the computation of fluid dynamic parameters, and the consideration of other possible effects on fluid flow and crystal growth under vertical, stabilizing and

microgravity conditions. An important aspect of the theoretical effort is the further development and improvement of transport models for diffusion limited mass transport of simple and of multi-component, multi-reaction vapor transport systems.

Progress to Date: Earlier accomplishments under this program include quantitative mass flux measurements and the analysis of crystal growth properties of bulk and layer-type crystals. These investigations demonstrate the effects of even minute fluid dynamic disturbances on the chemical and structural microhomogeneity of grown crystals.

Progress achieved during the present period of effort and on-going research are summarized below.

The dynamic microbalance technique, developed in our laboratory, was employed, for the first time, to the direct, in-situ, and quantitative determination of the Hg-vacancy concentrations and to the derivation of the enthalpies of vacancy formation in HgTe, $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$, and $\text{Hg}_{0.6}\text{Cd}_{0.4}\text{Te}$. More recently, these measurements were extended to the investigation of the metal vacancy formation in $\text{Hg}_{0.8}\text{Zn}_{0.2}\text{Te}$. The combined results reveal a slight but significant trend in the enthalpy of vacancy formation from HgTe to $\text{Hg}_{0.8}\text{Zn}_{0.2}\text{Te}$. These data provide the first, direct experimental evidence, in terms of vacancy formation energy, supporting theoretical predictions of the bond strengthening effect of Zn for this alloy system. The vacancy properties of binary and ternary compounds have a considerable influence on the electronic properties of these materials. Thus, the above investigations are of basic scientific and of technological significance for the further elucidation of the vacancy formation mechanism in semiconductor materials.

Primary emphasis was focused on the further development of experimental parameters for the USML-1 flight experiments of the $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ - HgI_2 vapor transport system. The stringent boundary conditions of the USML-1 Mission in terms of experimental time available present a considerable challenge for crystal growth experiments. In case of the epitaxial growth of $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ layers, the important parameters to optimize mass flux and growth rate are the temperature profile, source composition, and transport agent pressure. As a result of extensive investigations in our laboratory, the composition of the epitaxial layers is mainly controlled by the composition of the source material and by the transport agent pressure. The morphology of the epitaxial layers is strongly affected by the temperature and crystallographic orientation of the substrate. These results provide the basis for the definition of parameters for flight experiments. In addition, the results of several test experiments, performed in the Crystal Growth Furnace (CGF) Facility, are consistent with those obtained in our laboratory.

For the bulk growth of $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$, considerable progress was achieved in the development of a combined CVT-seeding technique in closed ampoules, which was not reported in the literature prior to our investigations. The objectives of this effort are to minimize multi-nucleation and to produce bulk single crystals by chemical vapor transport of sufficient size. The results of exploratory studies show that multi-nucleation is essentially absent and that high quality bulk crystals can be grown employing the above technique. Because of the technological implications of these results for the growth of $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ bulk crystals on ground and in space, emphasis is on the further development of the combined CVT-seeding technique for this system.

The continued theoretical and experimental investigations provide the scientific and technical basis for the performance and analysis of flight experiments of this system. In addition, the results of this work are generally applicable for the vapor phase crystal growth of ternary, alloy-type materials. Our previously developed predictive capability with respect to mass flux and crystal composition is of technological significance for the development, performance, and analysis of vapor phase crystal growth on earth and in space.

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2. SOLIDIFICATION OF METALS, ALLOYS, AND COMPOSITES

In-Situ Monitoring of Crystal Growth Using MEPHISTO

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October 1990 - October 1991

Objectives: The proposed experiments are being performed to gain a detailed understanding of the role of gravity driven convection during growth of faceted crystals. The scope of the investigation is defined such as to include the study of both the kinetics (i.e. the interfacial temperature vs. interface velocity relationships) and the morphological (i.e. the threshold and scale of interface instabilities and the resulting microstructures) aspects of solidification of faceted materials.

Research Task Description: A key aspect of the investigation is the use of non-invasive, direct and real-time monitoring of the s/l interface temperature as well as the demarcation of the interface shape during growth. This is achieved by using the Seebeck and Peltier techniques and an experimental configuration by which two constrained s/l interfaces are created in a cylindrical sample such that a central column of liquid is enclosed by a column of solid at each end.

Under static conditions (i.e. when the interfaces are stationary), both the interfaces are at T_m . However, when one of the interfaces is set in motion, its temperature deviates from the equilibrium value (interfacial supercooling), with the magnitude of the supercooling being dependent upon the imposed growth rate and the atomistics of the growth processes. The morphology of the moving interface is controlled by the local growth conditions such as the thermal and solutal fields and convective instabilities.

As mentioned above, for both the kinetic analysis and the detection of morphological instability (e.g. transition from planar to cellular) of the moving interface, the Seebeck thermoelectric effect is used: because of the temperature difference in the Seebeck coefficients of the solid and the liquid, a temperature difference between the two s/l interfaces (one stationary, the other moving) generates a proportional EMF which can be measured externally and non-intrusively. Using this approach, the atomistics of the interfacial processes (e.g. dislocation free vs. dislocation assisted growth) can be deduced from an analysis of the interface velocity vs. interface temperature relationship. For the morphological analysis, the Seebeck signal again provides a detection tool for the onset of instability, while the interface shape can be delineated by current pulsing, making use of the Peltier effect. Simultaneously, the growth rate is measured by monitoring the change in the resistivity of the sample as a function of time. Correlations between the interface kinetics, interface shape, microstructural morphology and microsegregation are then used to deduce the effect of thermosolutal convection.

In this program, pure Bi and dilute Bi-Sn alloys (both of which exhibit a highly facet forming tendency) are being used as model systems to generate an extensive database for growth under 1-g conditions: (i) to define the most pertinent conditions for experiments in the μ -g environment and (ii) subsequently for comparison with the μ -g results.

An integral part of the program is the extensive collaboration with the French MEPHISTO teams, both for the ground based and the space based experiments. As part of the collaboration, the various scientific, technical and engineering aspects of the investigation are being addressed concurrently at the University of Florida, CENG (Grenoble, France) and CNES (Toulouse, France). As part of an

agreement between NASA and CNES, the Bi-Sn experiments will be carried out on the second flight of the MEPHISTO hardware on USMP-2.

Progress To Date: Several important milestones have been achieved since the last report. These include: (1) tests of UF samples on the engineering model of MEPHISTO (Grenoble), (2) kinetic measurements in pure Bi and Bi-Sn alloys have been initiated at UF and at Grenoble (in collaboration with the UF team)- the early results indicate interfacial supercoolings on the order of 3 K at growth velocities of 0.8 to 1.2 mm/min, while interfacial breakdown at higher velocities has been detected using the Seebeck technique, (3) demarcation of the interface shape via Peltier current pulsing- to the best of our knowledge, this is the first instance where the s/l interface in dilute Bi-Sn alloys has been revealed by this technique, (4) determination of the Bi-rich side of the Bi-Sn phase diagram- resulting in new, more accurate values for the terminal solid solubility of Sn in Bi, liquidus slope and partition coefficient, (5) definition of process parameters for the growth of Bi-Sn single crystals- one single crystal has already been grown and valuable new information obtained on the morphological stability of Bi-Sn single crystal s/l interface as a function of growth velocity. Note that the ability to grow single crystals of required orientation is particularly desirable for kinetic as well as morphological studies.

In regards to the Seebeck technique, the preliminary kinetic measurements indicate that the total Seebeck signal comprises of a contribution not only from the s/l interface, but also from other, parasitic sources, commonly lumped together as "drift." For example, the experiments by the French MEPHISTO team on Sn-Bi alloys indicate that a large portion of the drift may come from sources such as grain boundaries. Consequently, our current experiments with single crystal samples will offer a concrete scientific basis for (a) identifying the sources of drift and (b) eliminating them, thereby improving the reliability of the kinetic data.

As part of the program, S.R. Coriell (Co.-I) of NIST is carrying out convective flow modelling and morphological stability analysis for Bi-Sn alloys. We are currently in the process of comparing our early experimental results with his predictions. A new set of tests is planned on the MEPHISTO engineering model (Grenoble) in the third week of November, 1991. Following these tests, UF samples will be tested also on the MEPHISTO flight model (Toulouse) in January, 1992. According to the current timeline, a project science review will be completed in March 1992.

Containerless Processing of Refractory Metals and Alloys

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Objectives: Research is being conducted on the containerless processing of refractory metals and alloys. There are three primary objectives of the work. One is to understand the kinetics of solid phase nucleation from refractory containerless melts. The second is to determine the growth velocity of the solid phase as a function of deep undercooling in refractory melts. The third is to relate the microstructure to the degree of undercooling and solidification velocity in refractory melts.

Research Task Description: The first of the research objectives involves both maximum undercooling measurements and nucleation frequency determination. Maximum undercooling simply requires conducting a set of experiments on a number of samples under the best possible conditions to obtain the highest possible undercooling. The effort on nucleation frequency requires repeated experiments on given samples to evaluate the distribution of nucleation temperatures as a function of cooling rate. The second objective involves monitoring the rate of recalescence in the samples and extracting the solidification velocity from that data. The solidification velocity is related to the degree of undercooling. The third objective is accomplished by postprocessing analysis on specimens for types, sizes, amounts, distribution and morphology of resulting phases. This includes detailing the distribution of elements and therefore characterizing segregation. A number of techniques for microstructural analysis are used including x-ray diffraction, optical microscopy, scanning electron microscopy with energy dispersive analysis, microprobe and analytical transmission electron microscopy.

Progress to Date: The investigation of the relationship between undercooling and solidification velocity in electromagnetically levitated melts has required application of innovative instrumentation capable of tracking the solid/liquid interface. The thermal field created by solidification was observed by using very high frame rate video imaging technology developed by Battelle Laboratories. The imaging system, capable of capturing 10,000 frames per second on a 64 by 64 pixel photodiode array, was used to observe solidification of Ti-51at% Al. These alloys were observed to undergo a solid state transformation from metastable beta to alpha. Solidification of pure nickel samples was also observed, but solidification occurred so rapidly to be recorded by the system. While the technology developed at Battelle provides much useful information it is limited to observation of solidification velocities from single nucleation points that are less than 20 ms⁻¹. In order to observe solidification of deeply undercooled melts and pure metals with solidification velocities greater than 20 ms⁻¹ a linear array of silicon photodiodes capable of 10 nanosecond time resolution has been implemented. This new development provides a means for both temperature and velocity measurement. It is being used to track the thermal field in undercooled nickel samples.

Pure zirconium samples have been processed in a containerless environment using electromagnetic levitation, induction heating, and optical pyrometry in order to measure the amount of undercooling achieved before solidification. Similar pure metal samples have been processed in the low gravity, containerless environment of the 105 meter drop tube located at the George C. Marshall Space Flight Center where optical pyrometry is also used to measure the nucleation temperature of the falling drops. Data acquired from both processing methods has been analyzed using statistical evaluation techniques which include the Kolmogorov-Smirnov test. These tests allow any differences to be detected between the two sets of data. The relevance of the observed similarities and differences is being investigated in terms of the specific characteristics of the individual processing techniques.

A series of niobium undercooling experiments were performed at CNES in Grenoble, France, in an ultra-high vacuum drop tube. The drop tube pyrometer developed by this group was used to record the undercoolings in some 21 experiments. No significant difference in undercooling between the Grenoble tube and the MSFC tube was observed.

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Alloy Undercooling Experiments in Microgravity Environment

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September 19, 1990 - December 18, 1991

Objectives: The objectives of this research are (1) to evaluate containerless melting and solidification of nickel and iron base alloys with and without softened glass coatings; (2) to develop techniques for the study of recalescence and growth behavior during solidification of undercooled alloy melts in microgravity; (3) to develop an understanding of undercooling phenomena in microgravity; and (4) to develop an understanding of the microstructures so produced.

Experimental techniques are being defined and documented for performing and analyzing undercooling experiments in space and on the ground. The scientific aim of the current modeling and experimental studies is to obtain baseline data and a baseline understanding of nucleation, growth, solidification, and structure of highly undercooled alloys on which comparable experiments will be conducted in space.

Research Task Description: The first alloy undercooling experiment in a microgravity environment was performed during the Columbia STS 61-C mission in January 1986. The results of the flight experiment have been published. Presently, science requirements are being defined and documented for a series of alloy undercooling experiments aboard IML-2 using TEMPUS hardware. The directly related ground-based experimental and analytical studies include thermal history measurements during the rapid recalescence and solidification of undercooled alloys, dendrite growth rate measurements, metallographic studies, and modeling of dendrite growth in alloy melts. Results of NASA sponsored work at MIT on solidification of undercooled alloys have been reported in about thirty published papers.

Progress to Date: Undercooling experiments have been performed using Ni-Sn, Fe-Ni, and Fe-metalloid alloys. New techniques have been developed for the study of undercooled melts in containerless Processing. A system for induction melting and solidification of iron and nickel base alloys coated with softened glass with simultaneous temperature measurement has been developed. High-speed cinematography has been utilized to observe the solidification of undercooled Ni-25%Sn alloy. Reheating curves measured during recalescence have been used to obtain dendrite tip velocity, fraction solid after recalescence, and other solidification features. Microstructure and microanalysis of samples produced were related to thermal and cinematographic data.

Recent work has emphasized techniques for levitation melting of droplets without softened glass coatings, for interrupted solidification studies, and for videotaping surfaces of samples during undercooling, recalescence, and final solidification. Samples can now be levitated in inert gas during heating and melting, and cooled by increasing the gas flow to induce undercooling. The sample can then be quenched, either before or after nucleation and recalescence, in a liquid gallium-indium bath at room temperature. This process allows the capture of microstructures representative of earlier stages of the solidification process in undercooled bulk samples than can be obtained by other techniques. It is also possible to drop an undercooled liquid sample on a solid substrate to initiate solidification and simultaneously to quench the sample with unidirectional heatflow. Appropriate substrate materials can be chosen to cause preferential nucleation of particular stable or metastable phases (e.g., either BCC or FCC in undercooled Fe-Ni and stainless steel melts).

The thermal data, video, and cinematographic data have provided direct insight into the mechanism of solidification of undercooled metal alloys. A detailed and comprehensive model is being

developed for processes occurring behind the dendrite tips during recalescence and subsequent solidification. The analysis includes evaluation of phase selection in nucleation and growth, heat flow during recalescence, solute redistribution during and after recalescence, and coarsening. The effects of these processes on grain refinement and microstructure morphology transitions are being examined and quantified using results from experiments on Ni-25% Sn alloy and Fe-Ni-Cr stainless steel alloys. A number of analytical techniques are being used to gather useful information about the solidified specimens, including x-ray and electron methods for determination of microstructural texture. This work will greatly enhance our understanding of solidification microstructures in undercooled alloys.

Experimental work on Fe-Ni alloys (5%, 10%, and 30% Ni) has confirmed the growth models developed from observations of the Ni-Sn system. Fe-Ni alloys are being studied, in part, because of the narrow equilibrium solidification range. At high undercoolings dendrite growth velocities are very high and nonequilibrium solute partitioning is an important consideration. In addition, in the Fe-Ni peritectic system a double recalescence phenomenon has been observed in alloys that normally would solidify with FCC austenite as the primary phase. Apparently, BCC delta ferrite nucleates preferentially, and the austenite nucleates after recalescence of the metastable delta phase. The same effect was confirmed by experiments using other iron base alloys.

Experiments have been performed with Fe-B, Fe-P, and Fe-Si alloys. These alloys are interesting because of their glass-forming ability, because of their very low partition ratios, and because of their wide freezing ranges. Observed dendrite growth velocities in these alloys have been measured, and are observed to be much slower than in the iron and nickel base alloys studied earlier. Recalescence times are generally one to three orders of magnitude slower. Recording of recalescence using a video camera in these alloys was shown to be feasible and more economical than film for measuring dendrite growth velocities. Detailed experimental data on recalescence in undercooled Fe-P alloys have been compared with recent theories of dendritic growth under conditions of nonequilibrium solute partitioning at the dendrite tips. Good evidence was found for a shift from equilibrium to nonequilibrium partitioning as undercooling increases.

An experimental investigation of the solidification behavior of several undercooled Fe-Ni-Cr stainless steel alloys has been initiated, using techniques described above. Alloys were chosen, based upon phase diagrams calculated using published thermodynamic data, in such a way that either BCC ferrite or FCC austenite may nucleate and grow as the primary phase at the onset of solidification, depending on alloy composition, undercooling, and substrate material (for quenched specimens). This work may help to elucidate the phase selection process in undercooled stainless steels. Prior work indicates that there is some disagreement among investigators, perhaps due to insufficient thermodynamic data and inaccurate phase diagrams. This study is aimed at clearing up some of the discrepancies in this technologically significant class of materials.

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Gravitational Role in Liquid Phase Sintering

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Objectives: This study combines ground-based and microgravity observations of liquid phase sintering to extract the gravitational role on both the macrostructure and microstructure levels. At the macrostructure level, the objective is to determine the factors leading to slumping, shape loss, and distortion during sintering. The microstructural level observations focus on grain size, grain size distribution, contiguity, connectivity, and crystallographic texture. In ground-based experiments with W-Ni-Fe and ZnO-BaO systems there have been extensive changes aligned with compact orientation with respect to gravity. Unfortunately, current theories are incomplete and can not handle the effects of particle contacts induced by gravity, while it is impossible to sinter compacts free of contacts in gravity to test theory. This research bridges both aspects of the problem by considering contiguity corrections to existing theories and by performing microgravity experiments free of gravity induced contacts.

Research Task Description: The primary focus has been on tungsten-nickel-iron compositions which exhibit classic liquid phase sintering behavior. The difference in density between the liquid and solid is nearly 9 g/cc, giving easily observed gravity effects. Measurements of microstructure evolution have been conducted during heating and during isothermal sintering for times up to 32 h. These examinations have been performed with two sample geometries and at three temperatures with five compositions. The resulting microstructural response information has been used to propose corrections to existing theories for liquid phase sintering. These corrections include the role of solid-liquid contacts induced by gravity. Considerable microstructural development occurs prior to the formation of the first liquid and liquid penetration of grain boundaries appears to be a key source of compact distortion during sintering. The long-term distortion is effectively a viscous flow process involving continual solution-precipitation of the rigid solid skeleton during sintering. Not only does the skeleton coarsen, but there is considerable grain rotation and the emergence of preferred orientation over time. In parallel experiments with ZnO-BaO, direct evidence of coalescence has been given. From these observations new heating schedules, atmospheres and processing sequences are being proposed for minimization of distortion and microstructural gradients in liquid phase sintered compacts. An upcoming Space Shuttle flight will provide critical samples with microstructures formed without the action of gravity. It is anticipated these will exhibit more uniform properties and will be free of solid-liquid segregation. Further, these samples will be the first sintered under conditions that are compatible with the fundamental assumptions of liquid phase sintering theory. Thus, they will provide a critical test of current models for grain growth, grain shape accommodation, coalescence, and liquid-solid separation.

Progress to Date: The effort to date has focused on developing a research protocol to clearly document the role of gravity at both the microstructural and macrostructural levels. Compacts have been sintered under a wide variety of conditions and analyzed for distortion, grain size, grain size distribution, connectivity, contiguity, grain orientation, and solid-liquid separation. These measurements have been used to carve out the critical conditions for flight experiments. Further, the observations have been used to test various theories. Past theories have failed to explain the results, leading to modifications that show agreement with the current observations. One of the most important aspects of the current work has been isolation and documentation of a clear gravitational role in liquid phase sintering. Current experiments are focused on answering important new questions that are not treated by existing theory. These are based on liquid phase sintering systems with similar thermodynamic and chemical attributes, but with differing ratios of solid density to liquid density. This will help quantify the role of density difference in solid-liquid separation during sintering. Additional experiments were performed on the ZnO-BaO model liquid phase sintering system to demonstrate the high level of grain coalescence during sintering, previously not treated by most models. Finally, in preparation for the IML-2 flight, test

compacts have been fabricated along with the needed crucibles. These have passed vibration testing and leak testing and will go into ground-based sintering on flight hardware in November 1991.

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Isothermal Dendritic Growth Experiment

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Objectives: Isothermal Dendritic Growth Experiments (IDGE) will be performed under microgravity conditions on high purity crystal growth systems in an apparatus mounted to the MPES carrier during the USMP-2 Shuttle spaceflight mission. Data from the IDGE, in which convection-free crystal growth occur, will provide a critical test of theory and add to our fundamental understanding of non-linear pattern dynamics by assessing the kinetics of unconstrained dendritic crystal growth in a convection-free environment.

Research Task Description: Dendritic crystallization is common in most industrial casting and welding processes, and it remains a subject of interest to both scientists and engineers attempting to understand the factors that control dendritic patterns and microstructures. Current theories of dendritic pattern formation are based primarily on diffusive transport, with convection either ignored altogether, or added in as a separate phenomenon. In actuality, the simultaneous presence of convection and diffusion during dendritic growth in the presence of Earth's acceleration field results in *coupled transport* that, in turn, alters the dendritic "operating state", which is often expressed as the speed and size scale of the crystals for a given supercooling, or thermal driving force. The ability to perform a critical test of dendritic growth theory depends on the availability of a suitable experimental system that provides precise, quantitative, thermodynamic driving forces acting on a well-characterized material.

The first IDGE flight, scheduled for the USMP-2 mission in October 1993, will measure dendritic growth events in succinonitrile (SCN: $\text{CN}-(\text{CH}_2)_2\text{-NC}$)-a transparent, body-centered-cubic plastic crystal that freezes with characteristics similar to the cubic metals. Procedures have been developed by the IDGE investigators to purify SCN to the extraordinary levels of purity ($>6\text{-}9\text{'s}$) needed to permit precise determination of the supercooling (melting point minus growth temperature) or free energy, available to promote dendritic growth.

Ground-based measurements have clearly delineated the influence of gravity on the dendritic growth process. Specifically, gravity starts to affect the growth rate and crystal morphology of SCN dendrites when the supercooling is less than about 1K, and buoyancy driven convection becomes an overwhelming kinetic factor as the supercooling decreases further. Buoyancy driven convection itself arises spontaneously from the interaction of the gravitational body force with the microscopic density gradients produced in the fluid phase by the thermal transport field responsible for the flow of latent heat from the crystal to the supercooled melt. At large supercoolings ($\gg 1\text{K}$) gravity plays a minor role, because convection is a weak transport process relative to thermal diffusion. However, it is only at small supercoolings, where gravity effects normally dominate, that the kinetic conditions are suitable for checking theories.

Thus, the first IDGE flight will perform photographic observations at a variety of supercoolings in the range 0.1–1.0 K. To accomplish this task a novel growth chamber, millikelvin thermostat, and optical system have been developed for operation under semi-autonomous conditions. The IDGE growth chamber permits a single sample of SCN to be repeatedly melted, supercooled, nucleated, and measured. Growth velocity and dendritic tip radius can be derived from the optical measurements, and these data then correlated with the supercooling. IDGE provides two independent optical axes for stereographic correction, and up to 250 35-mm photographic frames, which allow up to 20 independent dendritic growth sequences, at preselected supercoolings, each with a series of up to ten photographs taken for each millimeter of crystal growth.

On-board accelerometry-derived from the SAMS system-temperature measurement and control, and data for other relevant experimental conditions, including slow-scan video down-link of the dendritic growth events, are all available during flight in near real time to the POCC from the IDGE. Up-linking of a restricted range of commands for adjustment of experimental protocols, including experiment interruption, repeated runs, shift of supercooling, etc. are also communications features of the IDGE.

Progress to Date: During 1991, the P.I., his science team, and the NASA LeRC IDGE engineering team have concentrated their efforts on performing and analyzing integrated engineering tests of the IDGE prototype hardware. Methods for analyzing IDGE photographic data have been refined, including the development by the LeRC team of a novel digital image processing scheme for efficient IDGE photoanalysis. The new computer based digital method is currently being compared with standard optical techniques that rely on point-by-point image measurements made with a measuring microscope and an oscilloscope. The RPI science team is developing an improved process for achieving the reliable production of ultra-pure SCN based on a combination of high efficiency multiplate vacuum distillation with our standard zone refining methods. The new combined purification technique appears to reduce the total preparation time by almost 50%, and saves about one month in the time to prepare the SCN sample suitable for filling one IDGE flight chambers. IDGE chamber filling methods have also been improved and systematized over the past year, making that critical process more certain and repeatable.

Two significant science-related problems were identified during the initial integrated hardware testing include poor reliability of the temperature calibration scheme, using dynamic melting plateaus, and the lack of adequate image resolution in some of the IDGE photographs, especially in the range of supercooling 0.6-1.0 K. These problems are currently being addressed. An alternative calibration method for establishing the equilibrium melting point (liquidus temperature) of IDGE chambers has already been worked out using direct *in situ* microscopic observation of the solid-liquid interface within an IDGE flight chamber under slightly varying temperatures. As part of this effort we have also determined, by direct measurement, the pressure coefficient of the melting point of SCN, which is 22 mK/bar. This information, for which a consistent value was not available by calculation from thermodynamic data, is important, inasmuch as the IDGE growth chamber and thermostat operate at two atmospheres (absolute) pressure, and so a first-order pressure correction to the melting point is needed for setting signal levels within the temperature control computer software.

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Thermophysical Properties of Metallic Glasses and Undercooled Alloys

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Objectives: This project is aimed at studying the physical properties of undercooled metallic alloy melts which relate to glass formation. To accomplish this, we have proposed to develop calorimetric methods to investigate the specific heat and thermal conductivity of alloy melts both in the equilibrium and undercooled regime. The project includes use of conventional ground-based measurements (differential scanning calorimetry and drop calorimetry) together with the development of a noncontact calorimetry measurement on a liquid drop compatible with the microgravity environment of the TEMPUS electromagnetic positioning and heating hardware. The data obtained from these studies will be used in conjunction with a more general study of crystal nucleation kinetics in undercooled melts. In particular, we plan to investigate the degree to which refractory alloy melts can be undercooled. We will use the results of heat capacity measurements to determine the enthalpy and free energy functions of the undercooled melt, and will combine this with classical nucleation theory to develop a description of the nucleation kinetics and interfacial free energy between the melt and nucleating crystalline phases. Proposed alloy systems for study include the AuPbSb, AuGe, PdCuSi, NiZr, and NiNb systems (in order of increasing melting temperatures).

The results of heat capacity measurements will also be used to determine experimentally the entropy of a liquid alloy as a function of temperature and composition. By comparing this entropy function with that of the solid, the Kauzmann (and inverse Kauzmann) isentropic temperatures can be determined for solid/liquid equilibrium. The relationship of the Kauzmann temperature to the experimentally observed glass transition can then be assessed. It has been argued that the isentropic points for liquids and solids are associated with ultimate limits for undercooling and superheating, respectively. We plan to investigate this hypothesis.

Research Task Description: The project consists of two primary tasks. First, we will carry out a series of ground-based calorimetry experiments on several liquid metallic alloys. Using a SETARAM DSC 2000K and a Perkin Elmer DSC 4, we will determine the heat capacities of several glass-forming eutectic alloys as a function of temperature in both the liquid and solid phases over the broadest range of temperature experimentally achievable with this equipment. This includes measurements at lower temperatures performed with a modified DSC 4 operating in a liquid helium cooled cryostat. These low temperature data are essential for determining the total entropy function of solid phases. We are particularly interested in the difference in heat capacities of the solid and liquid phases as a function of temperature. This difference directly gives the rate of change of the entropy difference between the solid and liquid phases. These data can be used to evaluate the kinetics of crystallization in the undercooled alloy.

Our second task is to develop a noncontact specific heat measurement for high melting point liquid alloys which could be carried out in a microgravity environment using the TEMPUS facility. A systematic study of possible methods has been carried out. It was concluded that an AC method would be most suitable. An experimental AC technique which employs the TEMPUS electromagnetic positioning device, noncontact temperature measurement, and AC modulation of the power to the liquid drop has been developed. The concept is described in more detail below.

Finally, we are collaborating with the group of Eugene Trinh and Kinichi Ohsaka (of JPL) in their efforts to develop a ground-based technique for measuring the total enthalpy function of an alloy

melt in the undercooled regime. Ohsaka is currently utilizing an electromagnetic levitation system in our laboratory together with noncontact temperature measurement hardware developed at JPL.

Progress to Date: We have completed a detailed study of the crystallization kinetics in undercooled melts of the AuPbSb system. Measurements of the specific heat of the undercooled alloy were used to compute the entropy and free energy difference between the undercooled liquid and crystalline phases. These results represent the most systematic study to date of the thermodynamic function of a metallic glass forming alloy. The data in turn were used as direct input in an analysis of the nucleation kinetics of crystalline phases. We have also carried out measurements of the heat capacity of the solid and liquid phases of the eutectic alloy in the AuGe system using our Perkin Elmer DSC 4. We have measured the heat capacity of both the solid and liquid phases as a function of temperature. Data for the liquid phase have been obtained for 100°C undercooling. We have computed the entropy function for both phases and obtained an estimate of the Kauzmann temperature for the eutectic alloy. We have found that liquid alloys have both an excess negative enthalpy of mixing and an excess heat capacity with respect to the two phase solid alloy. We are in the process of measuring the specific heat of liquid AuGe far above the eutectic temperature (361°C) using our SETARAM DSC 2000K. These data are necessary in order to obtain an estimate of the upper isentropic temperature for this alloy.

As mentioned above, we have completed a developmental study of an AC noncontact calorimetry measurement to be carried out on the TEMPUS facility. The experiment employs the electromagnetic heating and positioning capability of TEMPUS. The AC method involves direct measurement of the AC temperature excursions of a liquid drop subject to and AC modulation in input power. The external relaxation of the drop temperature by radiation, the internal thermal relaxation of the drop, and other factors have been taken into account in designing an experiment. It has been found that heat capacity measurements with an accuracy of a few percent are feasible on a liquid drop with a radius ~ 1 cm at temperature in the range 1000°C - 2000°C. These conditions require an AC experiment at frequencies on the order of 0.1 Hz. In order to obtain absolute values of the heat capacity, it will also be necessary to determine the absolute power input to the drop as a function of its steady state temperature. We have determined that the best solution is to measure the total hemispherical emissivity of the drop (in both the liquid and solid state) in a ground-based experiment. With these data it will be possible to calibrate the power scale in the TEMPUS facility for each of our samples. We are working in collaboration with S. Krishnan of Intersonics, Inc. to measure the emissivities of our samples. Finally, as an added feature of the proposed experiment, one can determine the temperature dependent thermal conductivity of the liquid alloy. When the period of the power modulation is comparable to the internal relaxation time of the drop, the AC temperature excursions of the sample surface become frequency dependent. This frequency dependence can be analyzed to obtain the internal relaxation time of the drop. Under suitable conditions, this can be used to determine the thermal conductivity of the liquid alloy. As such our proposed experiment yields both heat capacity and thermal conductivity data.

Our collaboration with K. Ohsaka and E. Trinh has produced a drop calorimeter capable of measuring the total enthalpy function of a liquid alloy as a function of temperature, including the undercooled regime (see K. Ohsaka and E. Trinh, *Scripta Metall. Mat.* **25**, 1459 (1991)). We have used this apparatus and our SETARAM DSC to determine experimentally the heat of fusion and specific heats of the solid and liquid for a $\text{Ti}_{60}\text{Cr}_{40}$ alloy. Our interest in this system stems from reports in the literature that the metastable bcc solid solution (b-Ti) in this binary system transforms polymorphically to an amorphous phase upon annealing at 873 K (this crystal-to-amorphous transformation was dubbed "spontaneous vitrification" or SV). The existence of this transformation has been disputed in the literature by several investigators. We have used our specific heat and heat of fusion data to calculate the free energy difference, ΔG , between the liquid and the b phase as a function of temperature in the undercooled regime. Our results show that spontaneous vitrification of an undistorted b phase in the $\text{Ti}_{60}\text{Cr}_{40}$ alloy at 873 K is not possible because ΔG is positive at that temperature. However, ΔG may become negative with additional excess free energy to the b phase in the form of defects. This is in agreement with experiments that show that it is necessary to incorporate defects into the b phase before

spontaneous vitrification can be observed at 873 K. We are currently trying to produce an amorphous $\text{Ti}_{60}\text{Cr}_{40}$ alloy by sputtering so that we can attempt to measure the glass transition temperature and crystallization enthalpy. These data should improve our estimate of DG in the temperature range of interest (600 - 1000 K).

Publications

Fecht, H.J., "Thermodynamic Properties and Crystallization Kinetics at High Liquid Undercooling," *Proceedings of Workshop on Containerless Experimentation in Space* (E. H. Trinh, ed.), JPL Publication No. 9-30, 1990, p. 222.

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Casting and Solidification Technology (CAST)

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December 1986 - December 1991

Objectives: The objectives of research are: (1) to determine the influence of gravity on the fluid flow and nucleation that occurs during casting, and (2) to investigate the solidification and coarsening processes of dendrite arms and their subsequent influence on the grain structure in castings.

Research Task Description: The purpose of the investigation is to study the directional solidification of metal-model materials under low-gravity conditions. In particular, the inverted density layer and the thermal and solutal fields ahead of growing a interface will be analyzed. This investigation is an extension of previous low-g studies done on Space Processing Applications Rocket (SPAR) and KC-135 flights. To complete these studies, longer periods of low-g are required in order to allow solidification to occur at slower and more controllable rates. Therefore detailed analysis will be made of fluid motion near the solidification interfaces using the optical techniques (Schlieren, shadowgraph, interferometry and holography) available in the ground based laboratory and in the Fluid Experiment System on IML-1. To aid in separating the thermal and solutal profiles, thermocouples will be placed at intermittent locations along the cuvette wall. The temperature measurements will enable the investigators to mathematically separate the thermal and solutal effects on the interferograms.

Progress to Date: Two optics systems were assembled. One that provided confocal processing and particle tracking capabilities and a second interferometric system. The onset of convective instabilities within the inverted layer and concentration and temperature fields are being documented with this apparatus. A cuvette design that would accommodate shrinkage and expansion of the fluid was completed. An instrumented cuvette was assembled and operated under typical solidification conditions. Comparisons of concentration and temperature fields were made with optical and analytical predictions. A KC-135 flight rig was assembled incorporating a confocal optical system and VCR data recording. The advanced fluid thermal model has progressed continually and a graphics capability has been added so the solidification process can be viewed in real time as it is being calculated. Ground Control Runs have been completed on the FES hardware. The PI's are presently performing science runs in the holographic Ground System.

Publications

McCay, T.D., McCay, M.H., Hopkins, J.A., Lowry, S.A., and Smith, L.M., A Study of Convective Effects during Vertical Directional Solidification of Light Solute Rejecting Alloys," Int. Arch. Heat & Mass Transf., 1990 (accepted).

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Hopkins, J.A., McCay, T.D., and McCay, M.H., Interferometric Measurements of a Dendritic Growth Front Solutal Diffusion Layer," AIAA Paper 91-1334, *26th Thermophysics Conference*, Honolulu, June 1991.

Measurement of the Viscosity of Undercooled Melts Under the Conditions of Microgravity and Supporting MHD Calculations

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Objectives: The objective of this research is to develop the mathematical framework for planning an in-flight experiment involving the electromagnetic heating and positioning of an undercooled metal sample under microgravity conditions. The relaxation of the sample after the application of a brief current pulse which will generate an inward electromagnetic force that will squeeze the sample will be observed. The sample will relax in an oscillatory mode, and by examining the rate at which the amplitude of the oscillations is decreased, the viscosity of the undercooled melt can be deduced. In addition to providing a unique way of determining the viscosity of undercooled metallic melts, the project will provide information to other investigators performing undercooling and calorimetry experiments using levitation melting.

Research Task Description: The current research pursues three directions: (1) extensive computational work is being carried out to determine the effect of different materials and sample size on the electromagnetic force field, velocity field, temperature field, and free surface shape; (2) examine the behavior of deformed samples under TEMPUS experimental conditions; and (3) modeling of the deformation kinetics, the rate at which a sample is deformed upon the sudden application of an electromagnetic force field and the subsequent relaxation process.

Progress to Date: Important milestones of the research include the following: (1) the development of a numerical method to calculate the electromagnetic force field and predict the equilibrium shape of the metallic specimens; (2) calculation of melt velocities and temperatures in levitation-melted specimens; (3) calculation of what happens to the specimen, subject to both heating and positioning coils, when either the heating coil is switched off or when the current in the heating coil is reduced; and (4) confirmation of accuracy of free surface shape calculations by results of ground-based levitation experiments.

Publications

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3. FLUIDS, INTERFACES, AND TRANSPORT

Science and Technology of Surface-controlled Phenomena

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Dr. R. Glynn Holt
JPL Contract No. 958722
January 1, 1989 - June 15, 1993

Objective: The goals of this research are: (1) to determine the rheological properties of liquid drops in the presence or absence of surface active materials by exciting single drops into their quadrupole resonance and observing their free decay. The resulting data coupled with appropriate theory should enable us to understand better the physics of the underlying phenomena, providing a better foundation than earlier empirical results could; and (2) to investigate the mechanisms for coalescence of droplets with and without surfactants using a variety of techniques for perturbation of the interface between the drops. In addition to providing valuable physical insight into the coalescence process, practical knowledge in the form of determination of an energy-efficient approach to enhancing drop coalescence will be gained.

Research Task Description: The means by which we hope to achieve the aforementioned goals can be divided into two classes of effort: (1) ground-based efforts, both theoretical and experimental; and (2) microgravity experiments, to be performed on the inaugural flight of the Drop Physics Module on USML-1, scheduled for June, 1992. Our ground-based work has yielded insight into surface physics, acoustic scattering, drop dynamics, and acoustic drop control, and these results have served to guide and define the experiments on the DPM on USML-1. All of the tasks involve, both implicitly and explicitly, the interaction of liquid drops with acoustic fields. This interaction provides a containerless method for studying surface properties non-invasively, as well as (via resonant interactions with the shape modes of spherical drops) information about drop dynamics and interfacial properties for a time-dependent system.

Progress to Date: Single-axis levitation Experiments. The first project we will discuss is the single-axis levitation effort. Using an ultrasonic levitation apparatus we developed last year, we have made detailed measurements of the free quadrupole oscillations of water drops containing various types and concentrations of surfactants. The results of these measurements are contained in the article A.1. of the attached publication list, "Surface properties of surfactant-coated liquid drops acoustically levitated in air" of the attached publication list, along with a theoretical approach to the problem (see below, and article A.2.). Briefly, experimental results obtained thus far include: (1) decrease in frequency with increasing surfactant concentration; (2) increasing damping with increasing surfactant concentration. Combining these results with theoretical expressions obtained in A.1., we calculated (3) the decrease in surface tension, (4) the variation in Gibb's elasticity, and (5) the increase in the combined dilatational and shear surface viscosities with increasing surfactant concentration. In addition, we measured the variation of quadrupole resonance frequency with aspect ratio for different drop sizes. The observed decrease in frequency with increasing aspect ratio is not well described by existing theory, and thus stimulated further theoretical work (see below, and articles A.2. and A.3.).

Simulation of Drop Dynamics

To augment our understanding of the practical problems of positioning and manipulation in the DPM, experimental and computer simulation methods have been developed which allow ground-based investigation of the translational, rotational and vibrational motions of single and dual liquid drops in an external acoustic field in a microgravity environment. The acoustic fields used are the 3-D orthogonal resonant modes of a rectangular chamber similar to the Near Ambient Chamber. Our experiments utilize two resonant acoustic chambers, one providing a translation axis parallel to the gravity vector direction, the other affording an axis of translation in a constant g-potential plane. These chambers are acoustically

forced by a fully automated, software controlled system consisting of a microcomputer, A/D, D/A boards, plus external amplifiers and drivers. Using styrofoam spheres as sample levitation objects, we have designed and tested several successful schemes by which we can deploy, position, and bring together two samples in the chamber with a minimum relative velocity, facilitating our study of drop coalescence in air.

Since inertial effects can be expected to dominate the drop motion in the USML-1 environment, the relevant equations of motion were numerically solved on a computer which provides real-time graphical display of the results of changing acoustic parameters on drop motion for the large water drops scheduled for the DPM experiments. Since ground-based experiments cannot afford insight into such motions, the simulations are invaluable as a tool to develop and test various schemes for manipulation of the drops. As described in article A.4., "Simulation of drop dynamics in an acoustic positioning chamber", the most successful and robust of the translation schemes for dual-drop coalescence consists of the superposition of an even and odd mode of the translation axis, with varying relative amplitudes. Consideration of the acoustic force on an inclusion due to the two fields reveals a relative-amplitude-dependent equilibrium position where the resultant force is zero. By changing the relative amplitudes of the two modes, stable translation is achieved.

It should be noted that the resulting software package, designed to run on a Macintosh IIfx, is currently integrated into the USML-1 payload crew training procedures for DPM training at MSFC. The crew member is able to interactively alter acoustic parameters and immediately see their effect on the "drop" on the screen, thus developing familiarity and facility which will in all likelihood improve the science return of all the experiments scheduled for the DPM. Interested parties can obtain a copy of the software from our group upon request.

Drop Coalescence

We are currently performing experimental studies in hopes of shedding light on the fundamental processes involved in the inception phase of coalescence. The experimental scheme involves acoustic levitation of two immiscible liquid drops (usually hexane) in another host liquid (usually water) by exciting a typically mixed-mode resonance of a quartz rectangular chamber. The nearly-neutral buoyancy of the drops allows us to use a relatively gentle acoustic field to position the drops, thereby reducing distortion of the samples, and minimizing unwanted streaming effects. To investigate the hypothesis that coalescence in the presence of surfactants is induced by the action of capillary waves on the surface of one or both of the drops, measurements have been made in a system where cavitation occurs in the host fluid. High-speed video and camera records show in almost all cases of successful coalescence the presence of a cavitation event which triggers a capillary wave which traverses the inter-drop region, at which time coalescence occurs. A novel optical "cavitometer" has been developed to gather statistics on the relative efficacy with which cavitation events induce coalescence. Preliminary results indicate a time-dependence in the probability that a single cavitation event will induce a coalescence event which can be related to the spatial organization of the cavitation field and the surfactant concentration.

Theory

Surfactant-Coated Drop Oscillations

Analytical expressions have been developed for relating surface properties (elasticity, surface viscosities, surface concentrations) to the frequency and damping of free quadrupole-mode oscillations for water drops in air with surfactant dissolved inside the drop. This is done for purely spherical geometry. These expressions will meet their ultimate test when the results from our USML-1 experiments are available. However, they have been compared with the ground-based data in A.1., and used to obtain experimental estimates for the surface properties which are in line with our expectations.

Expressions are also developed which will, with the proper data, allow us to indirectly test some of the implicit assumptions of the model, such as the idealization of the equation of state for the surface concentration, and the diffusion of the surfactant species from the bulk to the surface.

Acoustic Radiation Pressure Effects on Drop Deformation and Location

In order to better account for the ground-based results, where ultrasonic levitation forces result in large static deformations of the drop equilibrium shape, and the force due to gravity causes the drop to be levitated below the position of the node of the acoustic field, the acoustic scattering of a plane standing wave from a spheroidal drop has been treated theoretically in A.2.. Assumptions of drop incompressibility and small deformations (i.e., constant volume and spherical symmetry) have been relaxed for our treatment, allowing us to obtain a self-consistent set of equations to be solved numerically for the unique combination of deformation and location which solves the problem for known external pressure and drop properties (size, composition). The results have caused us to schedule a set of experiments to simultaneously measure deformation and location vs. applied pressure and drop equilibrium size to test the theory. Comparison with previous researchers' data and theory (see the references in A.2.) shows that we can fit their data better with our new theory.

Oscillation Frequency Shifts

The eigenfrequency of the lowest order axisymmetric quadrupole mode oscillation will shift due to finite static and dynamic drop deformations in an acoustic field. In A.3., we employ a unique energy-based argument which does not rely on small deformations and spherical harmonic expansions to derive the oscillation frequency as a function of the drop deformations. We first treat the case for a simple shift due to a prolate or oblate deformation. Using results from A.2. for the deformation as a function of radiation pressure, we then include the effect of the presence of the acoustic field, and the time-dependence of the radiation pressure due to the change *during the oscillation* of the drop surface area. We thus show that it is possible, depending on the combination of acoustic and drop parameters, to obtain increasing or decreasing frequency shifts. For the particular set of parameters which match the experiments in A.1., we are able to fit the data for frequency vs. deformation for different drop sizes quite well.

Flight Experiment Definition

Determination of the exact sequence of experiments is complete. A long-duration single drop experiment is performed first, monitoring the long time scale changes in the surface properties in the NAC ambient environment by measuring the oscillation frequency and damping at regular time intervals over the 6-hour duration of the experiment. Second, experiments for single drop oscillations with varying concentrations of four surfactants (see attached final sample list) will be performed. Lastly, a series of dual-drop manipulation/coalescence experiments using both pure and SDS solution drops will be performed. Time estimates for all repetitions of the experiments have been made and approved. Imaging requirements have been tested on the DPM equipment at KSC during acoustic calibration tests. Ground-based analysis of sample purity, surface properties, storage properties and requirements, and safety considerations are all underway.

Publications

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Kinetics of Diffusional Droplet Growth in a Liquid/Liquid Two Phase System

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In-House

Objectives: The Chemistry and Polymeric Materials Branch conducts experimental and theoretical research to advance the science and technology of organic and polymeric materials having applications as optical and other technologically important devices. Studies relate to gravitational influences on solidification, crystal growth, thin film growth, and polymerization of such materials appropriate for study.

Research Task Description: Space laboratories in free fall, offer an environment in which body forces on fluids can be minimized over a relatively extended period of time and range of experimental conditions. Some advantages are to provide a quiescent environment to allow growth of more defect-free crystals, and solidification of more finely dispersed composites in low viscosity fluids by reducing buoyancy-driven convection and sedimentation. These effects are important, for example, in the study of factors yielding poor dispersions in miscibility-gap type system. A fundamental understanding of coarsening mechanisms, possible in low gravity experimentation, can lead to further understanding of factors contributing to fatigue in metal alloys, instability in foams, behavior of fogs and aerosols, and generally almost any of the multi-phase systems which cover a broad range of technologies. One such coarsening mechanism is Ostwald ripening. This is the process by which larger droplets grow at the expense of smaller ones by diffusion of mass away from droplets below a critical radius toward ones above this size. This branch is leading a research program funded to perform a flight experiment which will give data on the ripening process where gravity effects have been virtually eliminated. This experiment is designed to increase understanding of the fundamental phenomenon of diffusional grain/droplet growth.

Another research goal is to grow high quality thin films and single crystals of nonlinear optical organic materials in low-gravity by vapor transport, melt, and solution processes. Organic materials are of interest for use in all optical communication and signal processing systems because their nonlinearities are orders of magnitude larger than those of conventional inorganic materials. Additionally, organic materials offer flexibility of molecular design and a high damage resistance to optical radiation. They promise an unlimited number of crystalline structures, allowing optimization of device performance. Consequently, these materials might make it possible to replace electronic switching circuits in computing and telecommunication systems by purely optical devices. Microgravity should contribute to processing of high quality films and large single crystals of organic compounds, with optimum nonlinearities, nearly impossible on Earth due to convection.

Another branch activity related to droplet coarsening is examination of kinetics of the coarsening process due to droplet coalescence in an immiscible liquid-liquid dispersion in the absence and in the presence of gravity. The individual and collective roles of Brownian motion, Marangoni migration, and sedimentation on promoting coalescence. Current work is all ground-based with projected efforts toward establishing a flight experiment.

There is currently a feasibility study to apply Monodisperse Latex Reactor (MLR) technology to spherical dye-laser research. This activity has already successfully resulted in the insertion of fluorescent dyes into polystyrene spheres.

Progress to Date: The flight experiment to study Ostwald ripening in a liquid-liquid system is proceeding with ground-based testing. A method for deploying droplets of predetermined sizes has been designed. Testing of droplet deployment is in progress with evidence of good control. Computer

simulations of diffusion-driven separations provide a model, and will be used to determine the minimum droplet number to completely test the Lifshitz-Slyozov mathematical development. Simulation capability will impact hardware development cost by minimizing the need for hardware complexity.

Ground-based studies on organic crystal and thin-film growth have resulted in synthesis of a new pyrrole derivative of diacetylene with good third-order properties in acetone. Appropriate furnaces for thin-film and bulk crystal growth by vapor transport have been designed and fabricated. Single crystals of benzil have been grown by Bridgman and Czochralski techniques, and new polishing methods, suitable for organic crystals, developed. A healthy solution crystal growth program is underway with the goals of enhancing mechanical strength, by co-crystallizations, and developing viable middeck flight experiments. Several manuscripts are in preparation and several talks have been presented over the year.

Publications

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Interfacial Phenomena in Multilayered Fluid Systems

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NAG3-1094

Objectives: The objective of the proposed research is to investigate the flow structure of interface/surface tension gradient driven convection in a system of three immiscible liquid layers with two interfaces. This system is relevant to the encapsulated float zone and the Bridgman and Czochralski crystal growth techniques and also to dispersion material melts and groundwater pollution by hydrocarbons. Float zoning, and encapsulated float zoning is considered to be a qualified space processing technique. The incentive for investigating the phenomena under microgravity conditions is that primary forces which induce convection are interfacial forces while buoyancy forces are reduced. This provides an ideal opportunity for studying the interaction between two interface tension forces that either augment each other or compete with each other. A potential benefit of the study is suppression of surface tension driven flow in the encapsulated electronic material melt achieved in a low-g environment.

Research Task Description: The primary objective is to prepare a flight experiment on interfacial fluid mechanics for the IML-2 mission in 1994. The experiment is to be performed on the Bubble, Drop, and Particle Unit which is being developed by ESA.

Ground-based research is geared towards understanding the interaction of interface tension forces and gravity forces and their affects on the fluid physics of a multilayer fluid system. The light sheet technique is used to study the flow patterns. Holographic real time interferometry is used to evaluate temperature fields, and local thermocouples/RTDs are used to obtain information about any time-dependent structures in the flow. Test-cells of different geometries and end-wall boundary conditions are used for experimentation. Several experiments have been performed with such liquids as silicone oils, fluorinerts, ethylene glycol, hexadecane and water. Influence of the ratio of thermophysical properties on fluid flow is of particular interest. Also of interest is the influence of interface tension and the role of miscibility on fluid physics of multi-layered systems.

One dimensional models of the fluid flow provide a simple means of understanding the fluid physics. A one dimensional model is used to analyze a fluid mechanical system where buoyancy forces of variable strength may augment or oppose interfacial tension driven convection. Regions of distinct flow pattern are identified.

The case of a double layer system heated from below is studied to further understand the coupling mechanism between immiscible liquids. Linear stability analyses of the two layers heated from below are performed to determine the onset flow pattern of convection. Experimentally, it is observed that both layers are mechanically decoupled. While, one layer is in a conductive state the other may be convecting. When convection occurs in both layers, one may be steady state whereas the other is time-dependent. Shear flow develops at the interface without any significant effect on flow patterns in the layers, although the shear is presumed to be strongly dissipative. Flow directions of the convection cells appear to be developing independently in both layers.

Numerical flow simulation with the Navier Stokes equations provide predictions of the low-gravity flow patterns for the range of Prandtl numbers from liquid metals to oils. Single, double and triple layers have been studied. Attention has focused particularly on the time-dependent nature of these fluid systems at zero gravity. For moderate Prandtl number flows that are oscillatory at zero gravity,

terrestrial gravity exerts a stabilizing influence with respect to the time-dependent motion. For other moderate Prandtl number layered systems that are time-dependent at zero gravity, inclusion of buoyancy has been shown to elicit time-dependent motion presumably through return flow-thermocapillary interface interaction.

An analytical solution was proposed by Takao Doi of NASDA for infinite horizontal layers. Three "halt" conditions which stop the flow motion in the encapsulated layer were identified. A numerical experiment was performed to study the effects of vertical end walls on convection in an immiscible two layer liquid system ($Pr = 0.01$ over $Pr = 1.0$) in a two-dimensional cavity.

Using a Galerkin finite element technique (FIDAP), simulations were performed by J.P. Fontaine from ESA of the two layer immiscible system where the upper surface is a free surface. Both the top surface and the intermediate interface were considered deformable. This configuration is subjected to one-g and micro-g conditions. Micro-g conditions cause the strength of the flow in the lower layer to be greatly reduced. Also, a multicell flow structure is obtained in this layer.

Other significant issues for space flight experiments relate the stability of interfaces in a low-gravity environment. These issues are being investigated as part of a cooperative study between the Canadian Space Agency and NASA with the following objectives: 1) to identify the immiscible liquids that would have the most desirable characteristics for the flight experiment 2) to develop strategies that can be used to keep the liquids in the desired configurations throughout the flight experiment, and 3) to establish the interfacial tensions of the liquids and their temperature dependence. It has been observed that for a single layer a wetting angle lower than a critical angle of 35 leads to an unstable interface. These predictions were verified in zero-g simulation aboard KC-135 flights. Interestingly, the observations from the KC-135 flights are different to the results obtained in drop towers. The reason for this discrepancy is yet unclear.

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Pool Boiling Experiment

University of Michigan
Professor Herman Merte, Jr.
NAS3-25812 (NASA Contact: F. Chairamonte, LeRC)
March 1990 - September 1991

Objectives: This research is part of a program for the study of the fundamentals of nucleate pool boiling heat transfer under the microgravity conditions of space, seeking to improve the understanding of the basic processes that constitute boiling by removing the buoyancy effects which mask other phenomena, and which will be part of the development of data base for space applications of boiling.

The initial research focuses on the net forces acting on the growing/collapsing vapor bubbles when buoyancy has been drastically reducing, and consist of (a) internal vapor bubble pressure, (b) bulk liquid momentum, (c) molecular momentum of evaporation/condensation, (d) liquid viscosity, (e) liquid-solid-vapor surface tension, (f) Marangoni convection, and (g) residual buoyancy. It is desired to determine how these forces are related to the temperature distribution in the liquid, which itself is influenced by buoyancy.

Additionally, attempts will be made to describe the heterogeneous inception of boiling, using known liquid temperature distributions in the vicinity of the heater surface. This is possible in circumstances where nucleation takes place before distortion of the temperature distribution due to buoyancy occurs.

Research Task Description: A relatively large (19 mm x 38 mm) flat heat transfer surface is part of one wall of a closed vessel, maintained at a constant pressure and initially uniform temperature. The independent variables are subcooling and heat flux, with a step increase from zero to a prescribed power input. Measurements of surface temperature are made simultaneously with motion photography during the transient heating process including the onset of boiling, until a terminal condition is reached appropriate to the particular circumstances present. The heating surface used is a semitransparent layer of gold vacuum deposited on a quartz substrate, which acts simultaneously as a well-defined electrical heater and resistance thermometer, and which permits viewing simultaneously from the side and beneath the boiling surface. With heating taking place during microgravity the temperature distribution is precisely known at the onset of vapor bubble growth, as determined both visually from the photographs and from the accompanying drop in heater surface temperature. Both the spreading of the nucleation process at the heater surface and the motion and character of the liquid-vapor interface can be followed during the vapor bubble growth process.

Progress to Date: A total of 27 test drops were conducted in the 131 m vacuum drop tower facility at the NASA Lewis Research Center which provides 5 seconds of $a/g = 10^{-4}$, with 24 successful drops. These were conducted in support of the flight experiments, for which the Critical Design Review has been completed. A test vessel for the GAS experiment is under development, and an engineering model is currently being tested. It has been determined that heating times of more than 5 seconds will be required for six (6) of the nine (9) tests in the matrix.

Detailed analysis of the drop tower tests is continuing. Based on the tests conducted at $a/g = 10^{-4}$ and at $a/g = \pm 1$, six (6) distinct categories of boiling propagation across the heating surface have been identified to date. An explosive propagation with a striking pattern of small scale perturbances over the liquid-vapor interface occurred at relatively low levels of heat flux, giving rise to dynamic vapor bubble growths.

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Surface Tension Driven Convection

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Professor Simon Ostrach
Professor Y. Kamotani
NAG3-570 (NASA Contact: T.P. Jacobson, LeRC)

Objectives: The objective of the investigation is to design a thermocapillary experiment to study the transient and steady-state flows in the long-duration low-g environment of the Shuttle.

Research Task Description: The experiment consists of a circular container (5 cm dia. and 5 cm deep) filled with silicone oil, heating systems, and a data acquisition system. The fluid free surface will be heated locally by a CO₂ laser or by a submerged circular heater placed at the center. The resultant temperature variation along the free surface will generate thermocapillary flow in the container. The flow field will be studied by a flow visualization technique and the temperature distribution along the free surface, which is important because it determines the driving force of the flow, will be measured by a thermography technique. The surface heat flux distribution, the heating level, and the static free surface shape will be varied to study their effects on the nature and extent of the flows. Two series of experiments are planned. In the first one, the basic thermocapillary flow will be studied and attempts will be made to obtain oscillatory thermocapillary flow. In the second series the oscillation phenomenon will be studied in detail because it is considered to be an important aspect of thermocapillary flow. Ground-based and drop tower experiments together with a numerical analysis have been made to provide base data and to insure that the operating condition and the configuration will lead to flows that can be reasonably observed and measured.

Progress to Date: The experiment hardware was constructed and tested at LeRC and has been sent to KSC for integration to USML-1. A technique is being developed to measure free surface deformation in oscillatory thermocapillary flow in preparation for the future space experiments. A theory has been developed to explain the oscillation mechanisms and to show the importance of free surface deformation during oscillation.

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Thermocapillary Migration and Interactions of Bubbles and Drops

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Dr. R. Balasubramaniam, NASA Lewis Research Center
NAG3 1122 (NASA Contact: M. Hill, LeRC)

Objective: The objective of this investigation is to make experimental measurements of the thermocapillary migration velocities and shapes of single and interacting gas bubbles and liquid drops under the action of an applied temperature gradient in low gravity.

Research Task Description: The above experiments are planned to be conducted in the Bubble, Drop, and Particle Unit (BDPU) in orbit. The results will be compared with predictions from existing theory and new theoretical models to be developed as part of this research. The project is in the definition phase. The research task for the period involves performing tests on candidate systems and conducting feasibility studies, and providing consultation to the hardware design and development team.

Progress to Date: Candidate experiments were performed with air bubbles in silicone oils and methanol drops in silicone oils. Also, experiments were performed on the dissolution and growth of air bubbles in a silicone oil to assess the importance of size change effects. A draft Science Requirements Document was prepared. A computer program was prepared to permit calculations for estimating conditions for the flight experiments. Input was provided on a regular basis to ESTEC personnel regarding the science requirements needs for the experiment.

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USML-1 Drop Dynamics Experiments

Vanderbilt University
Professor Taylor G. Wang
Dr. Chun P. Lee
Dr. A.V. Anilkumar

Objectives: To study the shaping of liquid samples by acoustic forces. In particular, to examine the behavior of a liquid drop levitated and flattened in an intense acoustic field.

Research Task Description: The experiments are conducted in a free drop levitator at a fixed frequency of around 21.76 KHz. Water and glycerin are the typical drop liquids. Millimeter-sized drops are levitated and flattened with a slowly increasing input sound intensity from about 162 to 166 dB. The development of the drop shape is examined with high-speed video.

A relatively large water drop ($kR = 1$) changes from convex to concave shape as the sound intensity increases. Its liquid is squeezed to the rim that becomes a donut shaped ring, leaving a thin membrane at the center. Short wavelength ripples then appear on the membrane. The membrane then suddenly bulges upward while being flattened, with the thicker periphery contracting inward to form a closed shell that eventually shatters. For a smaller water drop ($kR = 0.5$), the ripples become unstable and emit satellite drops in both directions. An even smaller water drop ($kR = 0.25$) expands horizontally suddenly to disintegration after some moderate flattening. The details of this disintegration process are currently being examined. A glycerin drop does not display ripples and thereby does not atomize.

Progress to Date: From our theoretical studies, we believe that the ripples are capillary waves generated by the Faraday instability excited by sound vibration. Atomization occurs whenever the membrane becomes so thin that the vibration is intense. Buckling occurs whenever an existent equilibrium is unstable to radial motion of the membrane because of the Bernoulli effect. Also, the radiation stress at the rim of the drop is a suction stress which can make equilibrium impossible, leading to horizontal expansion and subsequent break-up of the drop.

Publications

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4. BIOTECHNOLOGY

Protein Crystal Growth in a Microgravity Environment

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Dr. Charles E. Bugg
NAS8-36611 (NASA Contact: K. Johnson, MSFC)
November 1990-October 1991

Objectives: The long-range objective for this research task is to develop systematic and reliable techniques and hardware for growing protein crystals in space. Studies will be performed to evaluate the potential for enhanced protein crystal growth under microgravity conditions. Fundamental studies of protein crystal growth, both on the ground and in space, will be performed in order to identify the major parameters that affect protein crystal growth. The fundamental studies will be used to define science requirements to be incorporated into advanced hardware that permit investigators to dynamically monitor and control the major parameters in protein crystal growth.

Research Task Description: This research program involves a multi-disciplinary effort to produce protein crystals in space of sufficient quality and size to permit molecular structural characterization by X-ray crystallography. The program simultaneously provides basic ground-based experimental and theoretical supporting research to develop a better understanding of protein crystal growth and to determine if gravity plays a limiting role in the growth process. Beginning with the Apollo program and extending into the Spacelab program, it has been demonstrated that microgravity can be used to enhance the size, morphology and internal order of protein crystals. A variety of proteins will be crystallized on shuttle flights during the next several years using advanced hardware developed at UAB that will permit real-time monitoring of the growth processes and will add a much higher level of automation. Our long-range plans include development of new methods for growing protein crystals, based upon the experimental and theoretical studies performed as part of this research program. A major goal is to develop advanced hardware that will be available for use on Space Station Freedom.

Progress to Date: Protein crystal growth hardware for use on the space shuttle was developed during the period between Challenger and STS-26. The hardware was constructed at Teledyne Brown Engineering Company, based upon the experience gained with four mid-deck flights in 1985 and 1986. The hardware includes a temperature control module, replaces a mid-deck locker, and contains three trays that permit 60 vapor diffusion experiments.

The hardware was flown in the mid-deck on shuttle flight STS-26 in September 1988. Three of the proteins included among the experiments on this flight produced crystals that were suitable for complete X-ray diffraction analysis. These three proteins were gamma-interferon, porcine elastase, and isocitrate lyase. The proteins had been studied extensively in crystal growth experiments on earth prior to the microgravity experiments. Analyses of the space-grown crystals included collection of three-dimensional intensity data sets with electronic area detector systems. Comparisons of the microgravity-grown crystals of these three proteins are larger, display more uniform morphologies, and yield diffraction data to significantly higher resolutions. Analyses of the three-dimensional data sets by relative-Wilson plots indicate that the space-grown crystals are more highly ordered at the molecular level than their earth-grown counterparts.

During the past year, the data obtained on STS-26 for porcine elastase and for gamma-interferon have been used in solving and refining the structures of these two proteins. The data for porcine elastase obtained from microgravity-grown crystals extend to 1.2A resolution, as compared to the previous resolution of 1.65A from earth-grown crystals. This enhanced resolution effectively doubles the data available for refining the structure of porcine elastase, and provides a large number of high-resolution structure factors for more precisely defining the atomic details of the structure. This structure has now

been completely refined using microgravity data to a conventional R-index of 16% at 1.2A resolution. The structure of gamma-interferon was recently determined using data obtained from crystals grown on STS-26, combined with the best data obtained from earth-grown crystals. This structure is now being refined at 2.75A resolution, with most of the high order terms obtained from one of the crystals grown on STS-26.

During 1991, protein crystal growth experiments were performed using the vapor diffusion apparatus on space shuttle flight STS-48. Since this flight occurred in September, the results from the flight are not yet available. However, this flight provided an opportunity to obtain detailed records of the activation and de-activation processes using a camcorder, and included 11 different proteins that should prove useful in further evaluating the effects of microgravity on the quality of protein crystals. In addition, protein crystal growth experiments were performed on STS-37 and STS-43 during 1991 using a new thermal gradient method for initiating protein crystal growth processes. Temperature change is expected to be a major new approach for future microgravity protein crystal growth experiments that involve the dynamic control systems to be developed during the next few years, and these preliminary thermal gradient experiments were designed to evaluate the potential of thermal change methods. The crystals obtained in these experiments were 20-30 times larger than crystals obtained in the control experiments that were performed in conjunction with the microgravity experiments. The protein used for these thermal gradient experiments was bovine insulin, which is a well-characterized protein. X-ray diffraction analyses are still in progress, but the preliminary data indicate that the microgravity-grown crystals from this thermal gradient system diffract to significantly higher resolutions than crystals from the control experiments.

Analyses of crystals from microgravity experiments performed in 1990 have now been completed. Crystals suitable for complete X-ray analysis were obtained of an antibody fragment on STS-31. Detailed X-ray analysis of these crystals, compared with crystals of comparable size from earth-control experiments, indicate that the antibody fragment crystals diffract to higher effective resolutions than crystals obtained on earth.

Along with the hardware development program, a number of basic studies in protein crystal growth have been completed. Particular emphasis has been placed on methods for detecting initial nucleation events, using laser light scattering and other optical techniques, and on methods for dynamic control of growth processes. Other work supported under this contract at the Georgia Institute of Technology has led to the development of an automated system for dynamic control of vapor diffusion rates. These studies will be combined to define hardware and techniques for dynamic control and monitoring of protein crystal growth parameters.

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Electrophoretic Separation of Cells and Particles From Rat Pituitary and Rat Spleen

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NAG 8-807

December 1, 1991 - November 30, 1992

Objectives: 1) To determine the effects of microgravity on the electrophoretic separation of rat pituitary growth hormone (GH) cell subpopulations; 2) to determine the effect of spaceflight on the ability of separated cells to synthesize and secrete biologically active GH; 3) to evaluate the capability of the FFEU (free flow electrophoresis unit) to separate GH particles containing unique 20K or 22K GH forms (or their polymers); 4) to determine if exposure of the GH cell to microgravity affects packaging of GH molecules within the secretory granule and 5) to determine effects of electrophoretically separated pituitary particles on immune cell function.

Research Task Description: Previous results from 3 spaceflight experiments (SL-3, COSMOS 1887 and COSMOS 2004) show that rat pituitary GH cell function is significantly reduced after exposure of the animal to μg for 7-14 days. Since the musculoskeletal and immune systems are GH targets, and since they are negatively affected by spaceflight, this issue warrants further study. We hypothesize that μg induced a change in the molecular packing density of GH within the secretory granule of certain GH cell subpopulations. We believe that the consequence of this change would be diminished hormone potency. By separating the biological particles in space, we not only test the various hypotheses, we also test the resolving capability of continuous flow electrophoresis techniques in μg .

Progress To Date: Cells. The function of cells in culture under various conditions of temperature, media, and substrate have been analyzed. The recovery of cells by enzymatic dispersion has been characterized, and the ability of culture media to maintain cells after extended storage has been evaluated. Results thus far show the feasibility of culturing pituitary cells in space for 5 days prior to harvesting, electrophoresis, and subsequent culture in μg . Particles. The requirements for a lysis fluid which maintains hormone in sedimentable form has been analyzed. A system consisting of water and zinc, which causes cell lysis in one minute, followed by the addition of a small amount of protease inhibitor results in quantitative recovery of immunoassayable hormone in the sedimentable fraction. Experiments have also demonstrated an absolute requirement for including deoxyribonuclease in the electrophoresis sample buffer. These results document procedural feasibility of this part of the IML-2 experiment.

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Protein Crystallization Experiments in Cryostat

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Professor Alexander McPherson

Objectives: The objective of the research project is to evaluate the characteristics and quality of protein crystals grown by the liquid-liquid diffusion method in microgravity. Specifically the experiments will utilize the Cryostat device designed and produced by DFRL and manifested for flight aboard IML-1 in January 1992.

Research Task Description: Prior to flight the Cryostat device is to be thoroughly tested for its ability to induce protein crystal growth under earth laboratory conditions employing specifically those proteins and solutions that will be utilized in the actual flight experiment. These include the proteins canavalin from the Jack Bean, catalase from bovine liver, and a plant virus, satellite tobacco mosaic virus. Improvements in the Cryostat device will be sought and any problems with the hardware are to be identified and corrected.

Progress to Date: A series of crystallization experiments have been carried out at both 22°C and 4°C using the proteins canavalin and catalase as well as with the virus STMV. These were carried out using the conditions intended for the flight experiment with variation of parameters, such as pH and precipitant concentration, around the mean values with the objective of optimization.

We have satisfied ourselves that the Cryostat device performs satisfactorily for the crystallization of all three types of samples under earth laboratory conditions. Crystals have reproducibly been obtained under a broad set of conditions and the optimal solution conditions have been identified.

The crystals that have been grown to date are morphologically identical to those grown by other methods, such as dialysis and vapor diffusion, in earth laboratory conditions. The quality and average size, though we have not accurately quantitated it, appears to be the same as those produced by the other methods as well.

In August 1991, the three Principal Investigators for Cryostat (Littke, Wagner and McPherson) met with Engineering Team Leader Peter Rank from Keyser-Threade and Maria Roth of DFRL for a three day training session in Freiburg, Germany. At this time all procedures for cleaning, loading and conducting the flight experiment were reviewed and mock exercises carried out. In addition, agreement was reached on procedures to be followed at Hanger L at KSC, transport of ground controls from KSC to the University of California at Riverside (UCR), and post flight analysis. Post flight procedures include removal of the samples from the flight hardware, transport from Edwards Air Force Base to UCR, and review of the crystals by optical techniques. Arrangements were also agreed upon for x-ray diffraction, data collection from space and earth grown crystals and subsequent processing. Currently we feel the experiment is in a ready state and fully ground tested.

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Enhanced Hybridoma Production Using Electrofusion under Microgravity

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NAG8-716 (NASA Contact: G. Spaulding, JSC)

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Objectives: Although newer, molecular techniques have now been introduced to prepare monoclonal antibodies, hybridoma technology remains critical to the production of monoclonal antibodies for use in a variety of biomedical applications. This technology relies upon immunological manipulation of lymphocytes, selection of suitable fusion partners, cell fusion and the selection of hybrids. All of these processes are, in turn, dependent upon a thorough understanding of cellular and subcellular processes underlying lymphocyte activation, regulation of the cell cycle, cell membrane biology and fusion. All of these processes are also central to many other biotechnological applications.

During the past year, we have extended our previous efforts directed at studying the biology of cell fusion and hybridoma production. The purpose of these studies has been to lay the groundwork for improvement of hybridoma technology, in general, and specifically, to provide a framework for the study of the effects of gravity on cell activation and cell fusion.

These studies have been focused on the following specific aims:

- 1) Mechanisms of lymphocyte activation and propagation
- 2) Mechanisms of immunological learning
- 3) Development and optimization of methods for the purification and analysis of cellular membranes of lymphocytes during activation and cell fusion
- 4) Evaluation and optimization of cell fusion experimental methods to obtain higher fusion yields and hybridoma recovery

Research Task Description and Progress:

1) Mechanisms of lymphocyte activation and propagation.

These studies have focused on both T-dependent and T-independent murine B cell activation. We have developed a model system of T cell-independent B cell activation that allows relatively long-term (14 day) propagation of certain murine B cell subpopulations, specifically B cells bearing relatively small amounts of the heat stable protein recognized by the monoclonal antibody, J11d (J11d^{lo} cells). These cells have been shown to be enriched for secondary B cell precursors. We have shown that the elimination of J11d^{hi} cells substantially increases the duration and extent of lipopolysaccharide/dextran sulfate mediated B cell proliferation. This observation has been exploited to produce murine hybridomas that are specific for a number of antigens. We have also shown that both J11d^{lo} and J11d^{hi} B cells may be activated in vitro using T helper cell lines.

We have also used model systems for B cell activation developed principally for space flight. The preponderance of evidence to date would suggest that lymphocytes may be unresponsive to stimulation *in vitro* by soluble activating agents, but may respond to cell-cell mediated activation. We have therefore developed a system based upon anti-T cell receptor monoclonal antibody activation and CD4⁺ T-B cell collaboration. We have found that this system efficiently activates and induces proliferation as well as differentiation of defined B cell subpopulations.

In addition to the initial cell-cell contact, a variety of regulatory substances may profoundly influence B cell responses. Various B cell subpopulations may differ substantially in their responses to such regulatory agents. To better understand factors influencing T-dependent B cell activation and to devise strategies for maximizing B cell activation, we examined the effect of cytokines on B cells activated using anti-CD3 bead stimulated CD4⁺ T cells. These studies showed that both interleukin 2 and interleukin 4 significantly increase immunoglobulin production by polyclonally activated B cells. In contrast, neurotransmitters such as Substance P, and Vasoactive Intestinal Peptide had either no effect on, or diminished immunoglobulin secretion (depending upon the source of the B cells used).

We are further evaluating the role of interleukins 5 and 6 and combinations of regulatory molecules in an attempt to optimize stimulatory conditions for B cell activation, proliferation and differentiation using both T-independent and T-dependent polyclonal activation models.

2) *Mechanisms of immunological learning*

One of the major goals of this project for the past year has been to establish conditions that maximize access to pure populations of fusible cells producing antibody with high affinity for preselected antigens. For this purpose, we have used the transfer of defined B and T cell subpopulations for the reconstitution of severe combined immunodeficiency (SCID) mice. This represents a preferred model system since, once conditions are maximized for murine cells, SCID mice, which themselves are devoid of an immune system, can also act as a vehicle for the stimulation of transferred human cells.

The production of high affinity antibody is dependent on the process of "immunologic learning" which accompanies the generation of "memory" B cells. "Immunologic learning" describes the increase in the quality (affinity) of antibody during the weeks following immunization. This is dependent on a dual process involving a) selective stimulation among the responding population of B cells with the highest affinity receptors for antigen, and b) the rapid accumulation of somatic mutations that change the specificity of responding cells, providing B cells with novel high affinity receptors. We have shown that the generation of memory B cells and somatic mutations in mice is relegated to a minority (10-13%) B cell subpopulation which can be enriched by virtue of its low expression of the cell surface antigen recognized by the J11D monoclonal antibody (J11D^{lo}). We have also shown that memory responses can be selectively regenerated in SCID mice by the transfer of J11D^{lo} precursor cells and T helper cells (T_H).

In order to fulfill the goals of this program we have assessed the generation of memory responses and immunologic learning in SCID mice reconstituted with J11D^{lo} precursor cells and T_H under various conditions. Our results to date have confirmed that this protocol reproducibly enables the generation of large numbers (2-8 X 10⁶) memory B cells specific for the immunizing antigen 2,4-dinitrophenyl-hemocyanin (DNP-Hy). Memory B cells can be found as early as 4 days after transfer and primary stimulation, and for several months thereafter. We have tested several antigen doses and found that responses are maximized at relatively high doses (250 µg).

We have now succeeded in measuring the affinity of numerous hybridoma antibodies obtained from SCID recipients secondarily immunized at various times after immunization. Our findings indicate that the average affinity of hybridoma antibodies generated by boosting 8 days after reconstitution and primary stimulation was 1 x 10⁶; whereas, the average affinity of antibodies for mice boosted at 39 days, when substantial learning should have occurred, was 2 x 10⁶. Thus, relatively little immunologic learning was observed. Therefore, we are attempting to increase the ultimate affinity of recovered antibodies by several strategies. These include: a) the use of suboptimal amounts of immunizing antigen, b) the use of passive antibody to decrease effective *in vivo* antigen concentration, c) the use of higher numbers of transferred T_H, and d) supplementation with other lymphoid cell subpopulations.

3) Development and optimization of methods for the purification and analysis of cellular membranes of lymphocytes during activation and cell fusion

Investigation of biomembrane structure associated with activation of T and B lymphocytes is essential for elucidation of the basic mechanisms responsible for normal functioning of the immune system. To facilitate biomembrane studies, improvements in basic separation and analysis technologies are required. During the past year we have concentrated our separation techniques on the use of free flow electrophoresis (FFE) to achieve our objectives. We have used the Hirschmann ACE 710 FFE instrument to document the usefulness of FFE as a research tool in monitoring biomembrane changes associated with electric pulse application (essential for electrofusion).

One difficulty encountered in these studies was the need for relatively large numbers of highly enriched cell populations from which membranes are prepared. This problem has been addressed by enriching for specific B cell subpopulations, activating and propagating them in sufficient numbers for membrane preparation using lipopolysaccharide and dextran sulfate. By flow cytometric analysis, we have shown that these cultures are greater than 90% enriched for J11d^{lo} B cells.

During the past year, our program has partially supported novel technology that permits separation of biomembranes with resolution comparable to that attainable with the Hirschmann apparatus, but is also capable of fractionation of preparative amounts of cell membranes as well as membranes of intracellular organelles. These studies have been done in collaboration with Dr. D. James Morré at Purdue University and with Dr. Ned B. Egen and Mr. Garland E. Twitty of the University of Arizona.

Further, additional concepts and strategies for membrane FFE separations have been developed and have resulted in disclosure of a new method and apparatus for amplifying peak to peak distance between sample-components (8) and is the basis for the Ph.D. work for R. Scharnez.

In addition, we have completed work on a computer-assisted program for image analysis of 2-D polyacrylamide gel patterns. These programs are important for the elucidation of the gene product changes associated B lymphocyte activation. Taken together, these studies have and will continue to allow us to advance our understanding of basic mechanisms and changes associated with the transition from the nonactivated to the activated state and the relationship of structure and function.

4) Evaluation and optimization of cell fusion experimental methods to obtain higher fusion yields and hybridoma recovery

The T cell independent B cell activation system for production of enriched populations of activated B cells has led to a reproducible source of fusible cells needed to optimize the electrofusion conditions. In order to obtain greater yields of hybridomas we have tested different combinations of electrofusion conditions and buffers. We are now applying these methods to human hybridoma production. In addition, we have begun a mathematical analysis of the forces engaged during alignment of cells during electrofusion. As a result it is now routinely possible to obtain yields of one hybridoma per 500-1,000 activated B lymphocytes. We believe that substantially further improvements in efficiency will be possible with additional refinements of these techniques.

Our ultimate objective is to obtain sub-populations of B cells specific to given antigens (see objective 2). These efforts have used T cell mediated activation approaches and were developed in parallel and funded independently through our NASA flight contract.

Because some B cell sub-populations represent a very small proportion of cells, a need for an instrument capable of fusing as few as 10,000 B cells was developed. This device is capable of yielding as many as 1,000 hybrid clones from a starting number of 10,000 fusible B cells. Future studies will

focus on the use of our newly acquired FFE methods, magnetic force separation methods, and flow cytometry methods to select for limited numbers of selected subpopulations of B cells as well as application to human hybridoma production.

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Electrophoresis Technology

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In-House

Objectives: The objectives of this program are to: (1) analyze the fluid flow and particle motions during continuous flow electrophoresis by experimentation and computation; (2) characterize and optimize electrophoretic separators and their operational parameters; (3) develop innovative methods to accomplish electrophoretic separations in space; and (4) analyze the electrophoretic process using apparatus that has been characterized or modified to perform in a predictable manner and according to procedures that have been developed to yield improved separation.

Research Task Description: Both horizontal and vertical laboratory electrophoresis test chambers have been built to test the basic premise of continuous flow electrophoresis that removal of buoyancy-induced thermal convection caused by axial and lateral temperature gradients will result in improved performance of these instruments in space. These gravity dependent phenomena disturb the rectilinear flow in the separation chamber when high voltage gradients and/or thick chambers are used, but distortion of the injected sample stream due to electrohydrodynamic effects causes major broadening of the separated bands observed in these chambers.

The electrophoresis separation process can be considered to be simple in concept but flows local to the sample filament produced by the applied electric field have not been considered. These electrohydrodynamical flows, formulated by G.I. Taylor in 1965 for drops suspended in various liquids, distort the sample stream and limit the separation. In addition, electroosmosis and viscous flow, which are inherent in the continuous flow electrophoresis device, combine to further disturb the process. Electroosmosis causes a flow in the chamber cross section which directly distorts the sample stream, while viscous flow causes a parabolic profile to develop in the flow plane. These flows distort the electrophoretic migration of sample by causing a varying residence time across the thickness of the chamber. Thus, sample constituents at the center plane will be in the electric field a different length of time and hence move more or less than comparable constituents closer to the chamber wall.

A moving wall concept has been developed for laboratory testing which will eliminate and/or control all of the above-mentioned disturbances. The moving wall will entrain the fluid to move as a rigid body and hence produce a constant residence time for all sample distributed across the chamber thickness. By aligning the moving wall at an angle to the chamber axis, a component of the moving wall motion can be made to oppose and hence cancel the electroosmotic flow. In the absence of electrokinetic effects, i.e. electroosmosis, the electrohydrodynamical effect manifests itself as a ribbon, being either vertical (perpendicular to the electric field) or horizontal (aligned with the electric field) depending on the ratio of conductivity of the sample to that of the buffer. Therefore, by using low conductivity sample solutions to provide a vertical ribbon, the moving wall concept should produce distortion-free separations.

A new electrophoresis-type test chamber has been built and put into operation. This chamber has the feature of two dimensional observation of the sample stream as well as observation of the sample stream cross section. The cross section is observed by means of a cross section illuminator which assures that the sample stream distortion can be accurately recorded.

Chamber wall zeta potential control is important in all types of electrophoretic separations. Experiments are currently underway to evaluate a published method to control electroosmosis by using

an external electric field. Several test chambers have been built to evaluate this control process in glass capillary tubes.

The separation of intact, chromosome-size DNA molecules is necessary to map and sequence the entire human genome. A method to accomplish this is being investigated using free fluid electrophoresis. A very high magnification microscope along with an associated test chamber is being set up to determine the orientation characteristics of DNA molecules under the influence of an AC electric field.

Progress to Date: Tests in an electrophoresis type flow chamber are currently underway to determine the effect of dielectric constant on sample stream distortion. This experiment work will settle the discrepancy which now exists between published experimental data and classical theory. This investigation will not only advance knowledge of the dielectric behavior of colloidal suspensions but also show how sample stream distortion must be treated in Continuous Flow Electrophoresis.

An investigation of electrokinetic sample spreading in zone electrophoresis is also currently underway. This work will use a numerical mathematical solution to completely model the buffer and sample system in a flow cross section. These numerical results will then be compared to experimental results obtained in a porous cellulose acetate film model of the flow cross section.

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5. GLASSES AND CERAMICS

Measurement of Liquid Interfacial Tension and the Role of Gravity in Phase Separation Kinetics of Fluid Glass Melts

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Objectives: The objectives of this flight program are: (1) To measure the liquid-liquid interfacial tension, as a function of composition, for a phase separating glass system. (2) To measure the phase separation kinetics of a glass which tends to micro-segregate on earth.

Research Task Description: Liquid-liquid interfacial surface tensions have not been measured by direct means for any immiscible glass systems. Since, however, the surface tension is one of the key parameters which enters into theories of phase separation kinetics, it is important to find procedures by which this quantity can be measured.

The liquid-liquid surface tension between end member compositions of a phase separating $\text{PbO-B}_2\text{O}_3$ glass will be measured in a containerless facility in microgravity. The measurement will be made by recording the distortion of a compound drop (consisting of the two end member compositions) which is subjected to rotation. The ratio of minor and major axes of the inner drop, when the compound drop is rotating at constant angular velocity, allows for the determination of the surface tension between the two liquids.

Progress to Date: During the past year the major effort has been devoted to the performance of property measurements on the organic liquids whose liquid-liquid surface tensions will be measured in space. The liquid-air surface tensions (at room temperature) of these liquids have been measured by the pendent drop method. Also, IR analyses have revealed that although the four liquid pairs selected for flight have been reported as nearly immiscible, certain of the components have non-negligible solubilities in their mated pairs. An experimental arrangement has been constructed which enables us to make liquid-liquid interfacial tension measurements by the pendent drop method. Extensive analyses of alternative schemes for measuring the drop dimensions have been performed. A procedure has been established which is accurate and yet not very time-consuming. Liquid-liquid interfacial measurements of the saturated liquid pairs have been performed, and we plan to report our findings in the near future.

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6. COMBUSTION SCIENCE

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Scientific Support for an Orbiter Middeck Experiment on Solid Surface Combustion

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Dr. Lin Tang
Professor Subrata Bhattacharjee (San Diego State University)
NAS3-23901 (NASA Contact: Kurt Sacksteder, LeRC)
December 19, 1984-December 20, 1992

Objectives: The overall objectives of the experiment are to: (1) determine the mechanism of gas-phase flame spread over solid fuel surfaces in the absence of any buoyancy induced or externally imposed gas-phase flow; and (2) improve the fire safety aspects of space travel.

Research Task Description: The spread of flame in the gas over the surface of a solid combustible involves in an essential way the transfer of heat from the flame to the solid fuel immediately ahead of it. This heat transfer is affected by the character of the gas-phase flow, and so the phenomenon of flame spreading under reduced gravity, in which the flow is generated by the gasification of the solid combustible and movement of the flame, is apt to be different from what occurs under the Earth's normal gravitational acceleration where the flow is largely buoyancy driven.

An experiment has been designed for the Middeck of the Space Shuttle to aid in understanding the process of flame spreading in the absence of a buoyancy-driven flow. A chamber of approximately 0.035 m^3 in volume is to contain either a thin sample of a cellulosic material or a thick sample of polymethyl-methacrylate and an oxidizing environment of O_2 and N_2 . Samples will be ignited at one end, and the ensuing flame spread process will be filmed. The spread rate can be determined from the films, and surface and gas-phase temperatures just above the surface will also be recorded. A matrix of eight experiments to be carried out on the Middeck has been identified. These data will help to clarify the mechanism of forward heat transfer in the low-gravity flames.

The experimental apparatus has been constructed at NASA's Lewis Research Center and tested in the Drop Tower facilities. Methods of data reduction and theoretical analyses of the reduced-gravity flame spread problem have been developed and are being refined. Results to date show that measured spread rates over thin cellulosic fuels are less at microgravity than for downward spread in normal gravity. Theoretically predicted spread rates not accounting for any effects of radiation heat transfer are generally higher than what is measured. Inclusion of surface reradiation in the modelling reduces the predicted spread rates and brings them more in line with experiment. Gas-phase radiation provides a feedback mechanism to the solid and a loss mechanism to the environment such that the effect of gas-phase radiation on the flame spread rate is rather complicated. Consideration of gas-phase radiation results in predicted spread rate trends that qualitatively compare favorably with experiment.

Progress To Date: Computational methods for determining gas-to-solid heat flux from experimental solid-surface temperature data for thin solid combustibles have been developed and applied to experimental drop tower results. The steady-state, computational model is now able to account for finite-rate gas-phase and solid-phase chemistry, surface reradiation, and gas-phase radiation. Both thermally thin and thermally thick fuels can be considered. Development of unsteady analyses to track the flame spread event from ignition to steady state or extinction is progressing, and some preliminary results have been obtained.

The first three matrix points of the SSCE experiment, ashless filter paper at 50% O_2 50% N_2 by volume at 1.5, 1.0, and 2.0 atm, were executed on STS 43 (7 October 1990), STS 40 SLS-1 (8 June 1991) and STS 43 (7 August 1991). Results from the experiment are now being examined, interpreted, and compared to theory. Increases in pressure result in increased soot production. Pyrolysis appears to be rather incomplete such that a substantial amount of potentially pyrolyzable fuel remains following passage of the gas-phase flame. Surface oxidation of the remaining fuel occurs to complete the experiment.

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Effects of Buoyancy on Turbulent Gas Jet Diffusion Flames

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July 1991 - October 1994

Objectives: The primary objective of this research is to improve the fundamental understanding of the influence of gravity on gas jet diffusion flames in the high-momentum laminar/ transition to turbulent/ fully developed turbulent regimes. Specifically, tests will be conducted in microgravity environments with the purpose of (1) isolation of buoyancy-induced turbulence by studying the characteristics of flame in the absence of this effect, and (2) characterization of the fundamental physical and chemical phenomena in turbulent combustion which may be masked by the presence of buoyancy.

Research Task Description: The basic mechanisms which control diffusion flames in particular and combustion in general include the coupled processes of mixing, chemical kinetics including soot formation and disposition, flame radiation, and buoyancy-induced convection. A major problem impeding our understanding of flame behavior is the dominating effect of buoyancy on flames that are studied in a normal-gravity environment. The behavior of fires on earth is determined by the combined convective and diffusive effects, where buoyancy-induced convection masks the processes of chemical-diffusional interaction that are fundamental to the understanding of combustion phenomena. In microgravity environments, the buoyant force is reduced substantially, and the remaining coupled processes become more tractable and easier to characterize.

There is an abundance of research results on turbulent flames; however, most deal with high-momentum type where buoyancy effect is negligible. This is not the case for low-momentum turbulent flames characteristic of unconfined fires. In this case, the fire-research community depends primarily on empirical results which, having been obtained under normal-gravity conditions, have the buoyancy effect inherently embedded within these correlations. Correlations of this type developed in a normal-gravity environment are not applicable to other environments such as those encountered in space, further exemplifying the need to quantitatively understand the effect of buoyancy on turbulent combustion.

There are two gravity-induced mechanisms responsible for the alteration of a low-momentum flow field. The first arises directly from the buoyant force acting on the time averaged or mean flow field and appears as a gravity term in the mean momentum equation. The second mechanism arises out of the interaction between density and velocity fluctuations which manifest in a source of turbulent kinetic energy. Under normal-gravity conditions, it is not possible to separate these two effects in terms of their impact on mixing rate, and hence, flame structure. Clearly, the advantage of operating in a microgravity environment would be to provide a major significant base of new information isolating the combined effects of buoyancy. Then, with a theoretical model, the effects of buoyancy on the mean flow and on the generation of turbulent kinetic energy would be separable. Isolation of buoyancy-generated turbulence will indeed provide data that will help improve our understanding of combustion and flow-generated turbulence. This makes the foundation for the current program, which provides critical data necessary to advance the understanding of turbulent reacting flows under both normal-gravity and microgravity conditions. As a result, the data will provide information from both fundamental and practical standpoints in relation to fire safety, practical combustion systems, and other aspects of turbulent combustion.

The program has just been initiated. This research will be accomplished in two phases, with definition (ground-based) period, Phase I, preceding the space-based experiment, Phase II. The primary purpose of the Phase I research is to conduct an extensive array of ground-based tests, associated analyses, and detailed numerical modeling. This phase will not only provide valuable and unique data on the behavior and characteristics of microgravity turbulent flames, but will define the science requirements, provide a concise and productive space-based test matrix, and help in developing the diagnostics for Phase II.

The ground-based, definition phase (Phase I) utilizes the 2.2-Second Drop Tower, the 5.18-Second Zero-Gravity Facility, and the KC-135 aircraft. Any diagnostics that may need longer than 5 seconds or are sensitive to the deceleration rates of the drop package will be tested and used in the KC-135 part of the experiments. Both qualitative and quantitative data will be obtained. Normal-gravity tests will be conducted to compare the results with corresponding microgravity studies. The 2.2-second tests will serve as a preliminary means of studying ignition, flame behavior, and fuel-flow rate, nozzle size, fuel type, and environmental requirements. Quantitative data will be obtained during the 5 seconds of low gravity in terms of temperature field, flame radiation, species concentration, chamber pressure, and visualization (cinematography). Also, thermophoretic soot sampling, light absorption/extinction for soot, velocimetry, Schlieren photography, and digital imaging with spectral interference filters will be examined. These are promising and feasible diagnostics for low-gravity experimentation.

Under the theoretical modeling task, the existing numerical models will be modified for application to turbulent gas jet diffusion flames. The modeling effort will incorporate multi-component diffusion (including thermophoresis), sub-global kinetics, radiation, and soot formation and oxidation. These submodels will be a part of a baseline closure formulation of the time-averaged equations. In addition, turbulence-chemistry and turbulence-radiation interactions will be included in further development of this model.

Progress to Date: To-date, extensive tests in normal gravity have been conducted to define the matrix for the ground-based program. Testing in the 2.2-Second Drop Tower is about to begin. Identification of submodels for incorporation in the numerical model has been initiated. The results of the 2.2-Second tests will define a matrix for the 5.18-second experiments.

The Science Requirements Document will be prepared by the end of 1992 for the planned (early 1993) Conceptual Design Review.

Publications

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Investigation of Laminar Jet Diffusion Flames in Microgravity: A Paradigm for Soot Processes in Turbulent Flames

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NAG3-1245

February 15, 1991 - September 30, 1991

Objectives: The objectives of this investigation are to study soot processes within steady and unsteady laminar jet flames in microgravity. The study is motivated by two factors: (1) laminar flame environments allow measurements of soot properties that are intractable in turbulent flames, and (2) soot processes in laminar and turbulent flames are closely related as long as differential-diffusion/buoyancy interactions of soot particles are not present.

Research Task Description: Project tasks include: (1) to conduct ground-based experiments, to gain a fundamental understanding of soot processes in nonbuoyant flames for a limited range of flame environments; (2) to develop a flight experiment, to provide results more relevant to practical turbulent flames that are not accessible using ground-based tests; and (3) to develop predictive capabilities for flame structure and soot processes, both to facilitate interpretation of the measurements and to provide information needed for computational combustion calculations of practical turbulent flames.

The ground-based experiments involve a short-drop free-fall facility that provides 1/2s in microgravity. To accommodate the limited test time, while providing reasonable soot concentrations, the jet flames operate at subatmospheric pressures within oxygen enriched environments. Measurements include distributions of soot volume and mixture fractions, temperatures, velocities and soot structure - all but the last involving nonintrusive optical diagnostics. Test conditions include various fuel types, ambient oxygen concentrations, pressures and flame residence times. These results will be used to address two main questions: (1) does the laminar flamelet concept apply to soot volume fractions, similar to other scalar properties, to provide a simplified approach for computing soot properties in turbulent flames; and (2) what is the effectiveness of detailed models of soot particle inception, growth, aggregation and oxidation that have been proposed in the literature?

The flight experiment will provide crucial information about soot properties in practical flame environments, in conjunction with the ground-based tests. Measurements will include soot volume fractions, temperatures (which can be related to mixture fractions) and soot structure, in the fuel-lean region of laminar jet flames in air at atmospheric pressure. The experimental results will be used to address the same questions as the ground-based experiments.

Theory will be used to interpret measured soot and flame properties, and to supplement the limited measurements of flame structure. The approach involves two-dimensional time-dependent computations of the laminar flames, exploiting the laminar flamelet concept to treat complex processes of fuel decomposition and soot chemistry for fuel rich conditions. These results then will be used to define the environment of soot particles for more detailed calculations of soot processes formulated in a Lagrangian manner (following the motion of the soot particles).

Progress to Date: Design of the ground-based test apparatus largely is completed and components currently are being fabricated and assembled. The test chamber is similar to the MICA configuration to be used for flight experiments. Effective gravity levels are less than $10^{-3}g$ due to drag on the chamber, which is satisfactory for testing at subatmospheric pressures. Current work on this phase of the study involves developing the suite of instrumentation for the experiments. It is anticipated that tests will begin early in the next report period and proceed continuously thereafter.

Work also was initiated to define the scientific requirements of the flight experiment. It was shown that light-emitting diodes provide an effective modulated source for extinction measurements to find soot volume fractions; and that establishing the stoichiometric contour of the flame from photographs was not feasible due to the intrusion of radiant emission from soot particles. Thus, flame structure will be defined from pulse-heated thermocouples (to control soot deposition) which are currently being developed. It is anticipated that the science requirements for the flight experiments will be defined early in the next report period.

Theory of the laminar jet flame structure has been developed, and a preliminary version (involving passive mixing) has been coded successfully and checked out on the computer. It is anticipated that development of this code will be completed early in the next report period, with subsequent work focussing on evaluating structure predictions and developing the more detailed analysis for soot properties.

Publications

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A Fundamental Study of Smoldering Combustion in Microgravity

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Michio Kitano
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NAG3-1252

Objectives: The objective of the overall research program is the design and performance of smolder combustion experiments under microgravity conditions, to help understand the mechanisms controlling smoldering, and in turn the prevention and control of smolder originated fires in normal gravity and in space-based environments. The specific objectives are: to develop ground-based experiments to determine the effect of gravity on the different modes of smoldering; to perform drop-tower and parabolic flight tests to obtain data on the smolder transition processes; and to use these data to design a space-based smoldering combustion experiment.

Research Task Description: The current research program is primarily experimental. Experiments are being conducted to determine the range of conditions for which smolder is affected by gravity, and how gravity affects the smolder process and its transition to flaming. The approach being followed is to measure the smolder velocity through materials of varied void fraction as a function of the velocity of a mixed (free or forced) oxidizer flow, for both downward and upward propagation. Comparison of the measurements for the two geometrical configurations allows the determination of the conditions where gravity affects smoldering. This information is then used toward the design of the space-based experiment.

The smolder process is too slow to obtain significant microgravity information in drop-tower, or parabolic flight experiments. However, such experiments can provide information about the microgravity behavior of the transition processes of smolder ignition, flaming and extinction. With this objective in mind, the present research task includes a series of drop-tower and parabolic flight tests aimed to obtain information about the suitability of the different ignition methods to initiate the smolder process in a microgravity environment.

Progress to Date:

1) Normal Gravity Experiments

Forced Flow:

A series of experiments of forced flow smoldering, opposed and forward, have been completed during this reporting period. In addition to providing information about the smolder process in forced flows, the experiments had the objective of determining the range of flow velocities at which buoyancy has a significant role in the smolder process. This was accomplished by comparing the results for upward and downward smoldering. Measurements conducted in these experiments included; the smolder propagation velocity and reaction zone temperature as a function of the air flow velocity, the location of the smolder front along the sample and the direction of smolder propagation (downward and upward).

The experimental results showed that there are some common characteristics to all the smolder configurations tested. All the experiments showed that three zones with distinct smolder characteristics can be identified along the foam sample. An initial zone near the igniter where the smolder process is influenced by heat from the igniter, an intermediate zone where smolder is self-sustained and free from external effects, and a third zone near the sample end that is strongly affected by convective currents. The smolder reaction propagation velocity and temperature generally have a direct correspondence and vary in each one of these three zones. The

analysis of these variations confirmed that the smolder process is controlled by the competition between the supply of oxidizer to the reaction zone and the loss of heat from the reaction zone. However, the variation of the smolder velocity and temperature with the forced flow is quantitatively different for opposed and forward smoldering due to the different effect that the flow direction of the oxidizer and post-combustion gases have in each case.

Opposed Smoldering:

The variation with the opposed forced air flow of the smolder propagation velocity and temperature shows that both parameters reach a maximum at flow velocities of approximately 2.5 mm/sec. At low flow velocities oxygen depletion is the dominant factor controlling the smolder process, and the smolder velocity and temperatures are small. Increasing the flow velocity strengthens the smolder reaction due to the addition of oxidizer, which results in larger smolder velocities and temperatures. At even larger flow velocities convective cooling becomes dominant causing the weakening and final extinction of the smolder reaction. These competing mechanisms play a very important role in the end region of the sample where buoyancy generated currents result in the strong enhancement of the reaction or in its extinction, depending on whether oxygen supply or convective cooling is the controlling smolder mechanism. Comparison between downward and upward smoldering indicates that gravity influences the smolder combustion of this type of foam for forced flow velocities smaller than 3 mm/sec, and sample sizes smaller than 50 mm.

Forward Smoldering:

The results of the dependence on the forced flow velocity of the smolder propagation velocity and temperature shows that in this case the smolder velocity always increases and the temperature decreases with the air flow rate, regardless of the sample location. These trends are the result of the hot postcombustion gases being convected ahead of the smolder front. Although they preheat the virgin material favoring the propagation of the reaction, they also dilute the oxidizer ahead of the reaction weakening it and reducing its temperature. For upward smoldering, transition to flaming was observed to occur in the char at the zone closer to the sample end and for air velocities of 15 mm/sec or larger. Transition to flaming did not occur in downward smoldering because the buoyant currents generated at the sample end oppose the forced flow. Comparison between upward and downward smoldering also showed that the effect of gravity takes place for air flow rates smaller than 3 mm/sec.

The differences observed between the opposed and forward smolder measurements can be explained by the differences in each case between the flow directions of the oxidizer and post-combustion gases. In the former case the cold oxidizer flows in the opposite direction than that of the smolder reaction propagation, and as the air velocity is increased, at low flow velocities, the addition of oxidizer dominates the convective cooling and results in an enhancement of the smolder reaction. At larger air velocities, however, convective cooling becomes dominant and the smolder velocity decreases as the air velocity is increased. In forward smoldering the oxidizer flows in the same direction as that of smolder propagation, and the hot post-combustion gases are convected to the virgin fuel ahead of the smolder front. The preheating of the virgin fuel results in an increase in the smolder velocity as the air flow rate increases even though the smolder temperature remains constant or even decreases due to the dilution of oxidizer by the post-combustion gases. Furthermore, the fresh oxidizer flows through the hot char, and since the char still contains a large amount of unburnt fuel, secondary reactions may occur in the char, which under certain flow conditions can result in transition to flaming.

2) Microgravity Experiments:

A series of smolder experiments were conducted at the NASA LeRC 2.2 secs drop tower to observe trends in the ignition characteristics of the foam, and to attempt to infer how smoldering will behave in microgravity. The parameter analyzed was the smolder reaction temperature variation with time, because the temperature itself, or the smolder velocity do not change enough in 2.2 secs to observe significant differences. The results for the temperature gradient variation with the flow velocity indicate that microgravity favors the initiation of smoldering, and that the upper range of flow velocities at which buoyancy plays a significant role on smoldering is around 2mm/sec, in approximate agreement with the normal gravity experiments.

A series of opposed flow smoldering experiments were also conducted in the KC-135 aircraft (30 secs of micro-g for up to 40 parabolas) to observe the effects of the variation of the gravity on the smolder process. Although the microgravity period was too short to study steady smoldering in microgravity, the tests provided initial information about the process and permitted the observation of smolder trends as the gravity changes. The tests also complement the Drop Tower tests summarized above. The results show that buoyancy affects both the species transport and transfer of heat to and from the reaction zone. At the reaction zone the former is dominant, which results in a decrease of the smolder temperature in microgravity. Away from the reaction zone the latter is dominant and the temperature increases due to the lack of convective cooling. All these effects are less noticeable as the flow velocity is increased, and as the reaction propagates toward the sample interior confirming that buoyancy is important at low flow velocities and near the sample ends. Another series of parabolic flight experiments have been conducted recently but the results have not been analyzed with enough detail to report them at this time.

Finally, the information obtained from the ground experiments on the smoldering of polyurethane foam has been used to design a small scale experiment to be carried out in the USML-1 mission scheduled for March 1992. The size of the fuel specimen (a cylinder 5cm in diameter and 10 cm long) is determined by the size of the Glove Box where the experiments will be conducted. Four tests are planned, two in still air and another two with a low velocity air flowing around the sample. Two igniter configurations will be used, one with the igniter at the cylinder axis and the other with the igniter at one end of the cylinder.

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Mechanisms of Combustion Limits in Premixed Gas Flames at Microgravity

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NAG3-1242 (NASA Contact: Dr. Karen Weiland, LeRC)
December 1988 - December 1991

Objective: The objective of this work is to gain understanding of the mechanisms responsible for limit phenomena in premixed gas combustion processes. By employing microgravity experiments it is possible to eliminate the complicating and sometimes ambiguous influences of buoyant convection on these phenomena.

Research Task Description: Three types of limit phenomena have been identified which may be affected by buoyant convection: flammability limits, ignition limits, and stability limits. The effects of the interactions of chemical reaction, thermal and mass diffusion, flame front curvature, and radiative transport are being examined. Additionally, newly discovered μg limit phenomena, not observable at earth gravity, are being investigated. These studies are compared with results of analytical and numerical models.

Progress to Date: We have found in previous work that lean H_2 -air mixtures exhibit unusual flame propagation modes at μg including cellular structures and in some cases stable, stationary, non-propagating spherical flames ("flame balls"). Our theoretical studies have shown that these result from the interactions of flame front curvature, gas radiation, and Lewis number (Le) effects. In recent experimental work, it has been found that the same behavior is found in lean H_2 - O_2 - CO_2 mixtures and H_2 - O_2 - SF_6 and CH_4 - O_2 - SF_6 mixtures of any stoichiometry. These results indicate that the Le of the mixture is the overriding factor in the mechanisms of these phenomena. Early studies were conducted in a drop tower providing only 2.2 seconds of μg , which was insufficient for studies of "flame balls." Recent μg experiments performed on NASA's KC-135A aircraft have shown that these structures appear to be stable for as long as low gravity persists. The g-jitter in the aircraft experiments caused a new phenomena to appear: "flame cylinders" resulting from moving "flame balls" which later break into "flame balls." While theory shows that steady "flame cylinders" are not possible, it also suggests that such structures can persist for relatively long periods of time.

In other work, we have studied the effects of radiation by adding inert, radiating particles to combustible gas mixtures. At low particle loadings, the particles increase the radiant energy loss, reducing burning rates and aggravating the tendency of these flames to extinguish. However, at high particle loadings, the mixture is optically thick, hence emitted radiation is reabsorbed within the mixture rather than lost to the walls of the combustion chamber. In this case, radiation enhances heat transport rates within the mixture. As a result, burning rates are found to increase to values *above those of particle-free mixtures*, in agreement with theoretical results.

Publications

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Abbud-Madrid, A. and Ronney, P.D., "Effects of Radiative and Diffusive Transport on Premixed Flames near Flammability Limits," in *Proceedings of Twenty Third International Symposium on Combustion*, The Combustion Institute, 1991, pp. 423-431.

Ignition and Flame Spread Across Liquid Pools

NASA Lewis Research Center

Dr. Howard D. Ross

Professor William A. Sirignano, University of California, Irvine

Objectives: The objective of this program is to determine experimentally the effects of gravity on the ignition susceptibility of and flame spread across liquid pool fires, and then to compare the results with predictions from numerical modelling being conducted at the University of California at Irvine.

Research Task Description: Experiments will be conducted in space to determine the ignition delay times and flame characteristics (steady vs. pulsating spread; flammability limits; flame spread rates; flame color; velocity and temperature fields) which will subsequently be compared to normal gravity experimental data and to two-dimensional model predictions.

Progress to Date: In the past year, drop tests were completed to determine the behavior of liquid pools in long, narrow rectangular channels, rather than the circular cylinders which have been used to date. It was determined that the liquid remained pinned in the cylinder, and that the characteristic damping time depended on the type of disturbance, i.e. the characteristic length changed depending on whether the disturbance was axial or width-wise. This information was required to better plan the space experiment.

Construction was completed for the 2 cm wide, 1 cm deep rectangular channel configuration of the experiments on flame spread over liquid pools being done in the Zero Gravity Facility. Additionally a low-speed fan was selected and installed to provide a crude forced flow (no flow straighteners). A short series of drop tests is planned to begin to examine ignition and flow effects. In addition, a new rectangular tray and cooling jacket was fabricated for use in normal gravity tests.

In the past year, the NASA grant continued to be supplemented by the receipt of 180 Cray-hours on the UCSD supercomputer. As such, both the open pool and enclosed pool problems could be studied. This year, most advances were made with the open pool, which emulates the experimental approach. The principal findings regarding preignition and flame spread processes as affected by gravity were: (a) gas phase buoyancy: Gas phase buoyancy carries heat away from the igniter in normal gravity, and thus reduces the amount of energy deposited on the liquid surface. However, while higher surface temperatures, and therefore higher vaporization rates, are consequently predicted in microgravity, the greater gas phase convection in normal gravity more rapidly carries fuel toward the ignition source, resulting in a prediction of slightly more rapid ignition in normal gravity; (b) chemical kinetics: the chemical kinetics algorithm, based on a split-operator scheme, was included in the code. Several difficulties were encountered, including the requirements of very short time steps and very fine spatial resolution. Nonetheless, ignition was observed, and the initial spread of a flame across a portion of the open pool was predicted. As expected, the leading edge of the flame is seen to lie in a premixed zone, while the trailing edge is a diffusion-type flame. Unfortunately, the flame is not yet self-sustaining: when the high-temperature igniter is deenergized, the flame extinguishes.

As a result of the work completed to date, it became apparent that space-based experiments are required to study ignition and flame spread involving deep pools. As such, a proposal was submitted to the NRA to continue portions of this work in a space-based laboratory. The proposal received a funding award in FY91 for flight project development, and work began on defining a specific set of space experiments. The Science Requirements Document was put in first-draft form, and transmitted to an engineering team for assessment. Finally, a new review article was drafted on ignition delays and flame spread behavior involving liquid fuel pools. It includes modelling results, flash point testing, recent experimental findings (post-1981, the time of the last review article), and little-known studies from Japan. It also highlights buoyancy-related processes and microgravity results.

Publications

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Scientific Support for a Proposed Space Shuttle Droplet Burning Experiment

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Professor Forman A. Williams

Princeton University

Professor Frederick L. Dryer

NAG 3-1081 (NASA Grant: Mr. John B. Haggard, Jr., LeRC)

October 3, 1989 - November 30, 1991

Objectives: The general objective of this program is to ascertain how best to utilize reduced gravity facilities to pursue scientific investigations of droplet combustion. The scientific objective is to provide scientific support for development of a droplet combustion experiment module for conducting experiments in the NASA LeRC microgravity facilities, and possibly aboard a space-based platform such as the Space Shuttle or in the Space Station. The planned experiments are intended to improve fundamental understanding of droplet combustion phenomena, especially in relationship to time-dependent characteristics and extinction phenomena. The scientific-support objective encompasses data acquisition and reduction as well as asymptotic and numerical modeling for interpretation of observations.

Research Task Description: The research tasks include theoretical modeling using asymptotic and numerical techniques, with emphasis on transient phenomena, detailed chemical kinetics and transport effects, ground-based experimentation on droplet burning in both the 2.2 and 5.5 second NASA LeRC drop towers, support to NASA in providing advice on hardware aspects of a flight experiment and analysis of data to be obtained from all experiments. The modeling addresses questions related to transient burning, to soot formation and destruction, to droplet disruption, and to ignition and extinction phenomena. Ground-based experiments have been focused on droplet ignition and on impulses imparted to droplets by ignition sparks; spark designs for minimum impulse were addressed. In addition, drop-tower experiments have addressed burning-rate measurements and mechanisms of soot production and of droplet disruption, as well as extinction phenomena. Work is encompassing n-decane, n-heptane and methanol as fuels in oxidizer/inert mixtures at pressures between about 0.2 and 2 atmospheres.

Progress to Date: In this work we have obtained data on the spherically symmetric burning of decane, heptane and methanol droplets in a range of ambient environments. The importance of the interaction between the spark and the droplet in the ignition of a droplet was established. Also, the importance of soot formation and how it influences the burning characteristics of hydrocarbon fuel droplets were demonstrated. Disruptive phenomena in droplet combustion were recorded and explained. The development of methods to employ a digital data processing technique for analyzing droplet combustion data was completed, and this was applied to droplet-burning data from drop towers. Identification of product dissolution in the liquid phase of a droplet was made, and clarification of histories of impurity buildup in the liquid during droplet combustion was achieved. It was found experimentally that the oxygen index limit for droplet combustion in microgravity is lower than in normal gravity.

In recent work, asymptotic analyses were completed for the burning rate and extinction condition of n-heptane droplets burning in atmospheres of mixtures of oxygen and nitrogen. Results were obtained for full chemical-kinetic mechanisms systematically reduced to two and three overall steps. Similarities of the flame structures to those of methane-air flames were established, but quantitatively significant differences were identified. A definition of scalar dissipation suitable for use in droplet burning was obtained, thereby providing a correspondence that can relate diffusion-flame structures and extinction conditions in different geometrical configurations.

Publications

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Presentations

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7. FUNDAMENTAL PHYSICS

Critical Fluid Light Scattering (Zeno)

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NAG3-849, December 15, 1987 - June 15, 1989
NAS3-25370, December 30, 1989 -
(NASA/Lewis Research Center, Dr. R. Lauver)

Objectives: The objective is to measure the decay rates of critical density fluctuations in a simple fluid (xenon) very near its liquid-vapor critical point using laser light scattering and photon correlation spectroscopy. Such experiments are severely limited on earth by the presence of gravity which causes large density gradients in the sample. The goal is to measure fluctuation decay rates with 1% precision two decades closer to the critical point than is possible on earth, with a temperature resolution of ± 3 microKelvin. This will require loading the sample to 0.1% of the critical density and taking data as close as 100 microKelvin to the critical temperature ($T_c = 289.72$ K). The minimum mission time of 100 hours will allow a complete range of temperature points to be covered, limited by the thermal response of the thermostat and correlation averaging times. Other technical problems have been addressed such as multiple scattering and the effect of wetting layers.

Research Task Description: We have demonstrated the ability to avoid multiple scattering by using a thin sample (100 microns), and a temperature history which can avoid wetting layers, a fast optical thermostat with microcomputer temperature control and measurement, and accurate sample loading. There remain the important engineering tasks of mounting the experiment to maintain alignment during flight and acceleration sensors to prevent Shuttle motions from contaminating the measurement data sets.

The experiment entails measurement of the scattering intensity fluctuation decay rate at two angles for each temperature and simultaneously recording the scattering intensities and sample turbidity (from the transmission). The analyzed intensity and turbidity data gives the correlation length at each temperature and locates the critical temperature.

The fluctuation decay rate data set from these measurements will provide a severe test of the generalized hydrodynamics theories of transport coefficients in the critical region. When compared to equivalent data from binary liquid critical mixtures they will test the universality of critical dynamics.

Progress to Date: During the current year the Zeno Engineering Development Model (EDM) was assembled, integrated and tested. The design shows fine temperature control, dual correlation function recording, satisfactory resolution of all noise issues from the flight design packaging, software capable of reliable remote, self-guided operation of the apparatus. Many hours of operation were done remotely using the serial port of the apparatus to connect over phone lines to the University labs.

The Critical Design Review was held in April 1991. At that time the crucial environmental tests (vibration and thermal) had not been completed. Much time and effort has gone into solving all the noise problems in the EDM and establishing a satisfactory optics mount design which will hold alignment to our tolerances. The sample cell design has demonstrated required performance in low leakage, cleanliness, and alignment. It now has sapphire windows, an integral valve, thermistor and platinum temperature probes. The optical alignment procedure has been demonstrated including full alignment of the thermostat with matched angles giving matched signals. The optical train (without thermostat) has demonstrated that it can hold alignment through launch. We are presently rebuilding a thermostat damaged in vibration tests and expect to finish the environmental tests on the EDM in October.

Publications

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Heat Capacity Measurements near the Lambda Point of Helium

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October 1, 1990 - September 30, 1991

Objective: Central to condensed matter physics is the phenomenon of second order phase transitions. These come about when a wide class of interactive terms are added to the simple ideal gas picture of matter. To understand condensed matter in general, it is necessary to address the phase transition issue, since these are involved in nearly all the interesting effects observed. Our goal is to perform the most stringent test currently feasible of the present theory of second order transitions in the asymptotic limit as the transition is approached. To do this we will measure the heat capacity of helium very close to its lambda transition at 2.1K.

Research Task Description: To perform the heat capacity measurements, two main requirements must be met: first we must have sufficient temperature resolution to establish the temperature scale, and second we need to control the energy input to the sample to determine its heat capacity. To these ends we have been developing a new high resolution thermometer and an advanced, multi-layer thermal control system. The thermometer makes use of superconducting technology to achieve a resolution of about 3×10^{-10} deg in a 1 Hz bandwidth, and the thermal control system can achieve a power resolution approaching 10^{-12} W. These two systems give us the capability to make measurements to the limits imposed by the Shuttle environment. A third requirement is to achieve an operating temperature of 2.1 K. To do this we make use of the superfluid helium research facility previously flown on Spacelab-2 by JPL.

Progress to Date: In mid 1990 the flight instrument electronics was shipped to JPL for integration with the low temperature research facility and subsequent environmental testing. In the last part of 1990, some delays encountered due to low temperature leaks in the dewar, but by the end of the year we were ready to commence testing of the integrated system. Performance testing was successful and the flight system was subjected to the environmental test program. A number of small problems were encountered and solutions were implemented. The system is now entering the final functional test in preparation for the pre-ship review. We expect to ship to KSC in January 1992 in preparation for a September 1992 launch as part of the USMP-1 payload.

Publications

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Israelsson, U., "The Lambda Point Experiment at JPL - A Status Report," in *Proceedings of the Eighth Oregon Conference on Low Temperature Physics*, January 1991, p. 74.

Critical Fluid Thermal Equilibration

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Dr. R. W. Gammon, University of Maryland
Dr. R. F. Berg, NIST (Gaithersburg)
Dr. M. R. Moldover, NIST (Gaithersburg)
Dr. J. Straub, Technical University of Munich (FRG)

Objectives: This experiment will examine the thermal relaxation and the fluid density profile as a function of time after a temperature perturbation of sulfur hexafluoride (SF₆) near its liquid-vapor critical point in the low-gravity environment of the Shuttle. Past low-g critical fluid experiments yielded unexpected results which were perhaps due to unanticipated time scales for relaxation dynamics. Future critical fluid experiments will depend on achieving thermal equilibrium to within a specified tolerance and on knowing how phases develop or disappear. This work is intended to determine the practical time scale needed to execute meaningful critical fluid space experiments and characterize the location and dynamics of density or phase domains within the sample.

Research Task Description: The experiment time-line allows observation of large phase domain homogenization without and with stirring after thermostat installation in the Critical Point Facility (CPF), time evolution of heat and mass after a temperature step applied to one-phase equilibrium sample, phase evolution and configuration upon going two-phase from one-phase equilibrium, effects of stirring on a low-g two-phase configuration, two-phase to one-phase healing dynamics starting from a two-phase low-g configuration, and quantifying the mass and thermal homogenization time constant of a one-phase system under logarithmic temperature steps. Two Critical Fluid Thermal Equilibration (CFTE) test cells will be developed to be flown in the European Space Agency's CPF, which is manifested on IML-1 for January 1992 launch. During the full experiment, accelerometry data, time correlated with the video records, will identify the compressible fluid dynamics associated with Shuttle acceleration events and provide the investigators with intuition about gravity effects in a non-vibration isolated Shuttle environment.

Progress to Date: The delay of the flight from December 1990 to January 1992 has slowed science progress. The year has been spent redoing experiment time-lines to accommodate a 2 day shorter mission for IML-1, re-verifying the software time-line in the engineering model, recalibrating the temperature scale of the flight hardware, and reviewing several mission time-lines for consistency with the science time-line and condensing them for science operations during the mission. Performed graphical analysis of thermal behavior of the CPF hardware to be better prepared to understand expected gradients on the fluid. Executed image processing on several video interferogram images generated by the hardware to find the best quality source. It turns out the raw digital data stream from the Shuttle provides the cleanest image source, but it is the hardest to process. By the end of October 1991 the investigator team will have completed three weeks of mission operations/training with two more weeks to go before launch. No publications in this area were produced by the investigators.

C. ADVANCED TECHNOLOGY DEVELOPMENT (ATD)

Chemical Vapor Deposition Facility for Reactor Characterization (CVDF)

NASA Langley Research Center
Dr. Ivan O. Clark
In-House

Objectives: The objectives of this research are to enhance the basic science of fluid dynamics for non-isothermal reactive fluid flow and to apply the unique laser velocimetry system of CVDF to support the overall microgravity program.

Research Project Description: This microgravity science program at Langley Research Center is centered on understanding the reactive fluid dynamic flows found in chemical vapor deposition (CVD) reactors. Various forms of CVD are used for manufacturing technologically important films in semiconductors, optics, fiberoptics, and wear- and corrosion-resistant coatings. The quality and uniformity of thickness and alloy composition of the layers formed by CVD are tightly coupled to the flow of gasses in the reactor and to the heat and mass transfer in the region of the deposition. This is a research area in which increased scientific understanding can be rapidly translated to improved industrial processing.

The use of microgravity is very important for developing a quantitative understanding of the forces involved in reactive fluid flows. On Earth, unavoidable horizontal thermal gradients in these flows result in very strong buoyant convection. The velocities arising from this thermally driven convection are frequently larger in magnitude than those due to the external forced convection. Hence, it is not generally possible to resolve the effects of solutal buoyant convection and of forced convection due to thermal expansion, reaction volume change, and Soret diffusion from the dominating thermal buoyant convection. By performing experiments in microgravity, the thermal buoyant convection will be greatly reduced and the other convective forces can then be systematically investigated.

Both experimental and theoretical approaches are pursued at Langley Research Center. For the experimental research, a three-dimensional laser velocimetry (LV) system is used to measure the flow velocities in CVD reactors. This system represents a technology transfer from NASA's aeronautical research to the field of semiconductor research and presents an unique measurement capability for the electronics industry. The capabilities of the facility have been tested and demonstrated through collaborations with Research Triangle Institute and ITT. Results to date have served to demonstrate the dominance of the thermal buoyant convection at unigravity as well as develop a better understanding for the design and operation of these reactors.

For the theoretical research, numerical modeling tools are being validated, and applied to understanding reactive fluid flows. Under support from the Small Business Innovative Research program, numerical modeling tools are being developed using both finite difference and spectral element techniques. As part of the microgravity research effort these codes are being validated and applied to current research problems. The finite difference code was used, with good results, to model the deposition of gallium arsenide (GaAs) by metalorganic chemical vapor deposition (MOCVD) in an horizontal CVD reactor. Current efforts in this area focus on studying the MOCVD growth of indium phosphide in the same horizontal CVD reactor as used in the GaAs effort. This code is also used to model a co-flowing laminar diffusion flame in support of experimental drop-tower research at Lewis Research Center. Due to the similarities in reacting flows for combustion and deposition, this has proven to be a good test of the capabilities of the numerical code. The wider range of reaction rates found in the combustion experiment has proven to be very challenging for the numerical model, but very good results have now been obtained. The numerical modeling tools are also being used to model experiments for the Advanced Automated Directional Solidification Furnace (AADSF). This effort is in support of AADSF characterization and flight research experiments.

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Advanced Technology Development (ATD) in Interface Measurements

NASA Langley Research Center

Dr. A.L. Fripp

Mr. W. Debnam, NASA Langley Research Center

Dr. P. Barber, Longwood College

Dr. D. Schleich, Polytechnic University, Brooklyn

Objectives: The objective of this ATD is to develop techniques to measure the shape and position of the liquid-solid interface in crystal growth.

Research Task Description: The interface position is determined by furnace conditions and the sample thermophysical properties. The interface shape is interactive with its position in the furnace and the mass transport in the liquid. The measurement of these parameters is important to both controlling defect formation in the crystal and to the scientific understanding of the experiment.

The approach to develop this measurement capability is two fold. The technique that has been most highly developed is to observe the interface with x or gamma radiography. The other technique is to use ultrasonic waves to characterize the interface.

The radiographic measurement technique has been remarkably successful. Experiments have been conducted in which the shape and position of the interface were observed during the entire run. The interface was observed to move, relative to the furnace, toward the hot zone during the run when the melt did not change composition as a function of growth time. In runs where the melt lost the higher melting point solute during growth the interface moved toward the cold zone. In no case did the actual growth rate equal the ampoule pull rate. The shape of the interface changed from convex to flat to concave as the run progressed.

Measurement of the solid-liquid interface position during Bridgman growth has been measured using ultrasonic waves. Although this technique is not as well developed as the radiography technique it is still a promising area and may be more integrateable into flight hardware.

The ability to observe this phenomena, in real time, without disturbing the growth process will greatly enhance our scientific knowledge of crystal growth.

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Vibration Isolation Technology (ATD)

NASA, Lewis Research Center
Joseph F. Lubomski

Objective: The objective of the Vibration Isolation Technology Project (ATD) is to provide the technology for the isolation of microgravity science experiments by developing methods to maintain a predictable, well characterized and reproducible low gravity environment consistent with the science requirements of microgravity science experiments.

Research Task Description: The Vibration Isolation Technology Project (ATD) is being conducted in three elements or phases:

- _ Definition of technology requirements
- _ Technology development
- _ Technology demonstration

The definition of technology requirements is being accomplished by assessing science requirements for selected microgravity science experiments and by analytical modelling to determine the sensitivity of selected science experiments. The latter effort is being conducted and directed by the NASA Marshall Space Flight Center members of the VIT Project. The technology development phase is being conducted in-house at LeRC and through university grants. This phase is presently concentrating on low frequency actuator development and the associated control technologies. Hardware is being developed for laboratory evaluation of concepts and principles. The technology demonstration phase is an in-house effort consisting of proof of concept demonstrations in a laboratory and then a systems demonstration in a low gravity environment using the LeRC Learjet aircraft. A six degree-of-freedom actively controlled, magnetic isolation system is being developed, and will be flown, for proof of concept in the technology development phase.

Progress to Date: A Passive Isolation System Testbed and Data Acquisition System were flown on the Learjet in November of 1990 as part of the technology demonstration phase. Five flights with a total of 26 low gravity trajectories were flown. The purpose of these flights were to qualify a Testbed and Data Acquisition System for proof of concept demonstrations using the Learjet later in the project. In March of 1991 a six degree-of-freedom, actively controlled, magnetic isolation system was successfully demonstrated by closing the loop around a position sensor and an inertial sensor. An isolation capability of 120 dB per decade was attained. Isolation down to 1 micro-g was attained which is the sensor noise floor. An International Workshop on Vibration Isolation Technology for Microgravity Science Applications was conducted in Cleveland, Ohio in April 1991. Workshop proceedings are being prepared for publication. A six degree-of-freedom actively controlled magnetic isolation testbed system was designed, built and integrated into a Learjet rack. Proof of concept flight testing of the actively controlled magnetic isolation system will be conducted in early October 1991. The ATD annual review was conducted at NASA Headquarters in July 1991.

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Laser Light Scattering Instrument

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In-House

Objectives: The overall objective of this Advanced Technology Development (ATD) project is to provide a Laser Light Scattering (LLS) Instrument for use in a microgravity environment. This entails an assessment of user requirements, an exploration of the capabilities of existing and prospective laser light scattering hardware, and a coordination of the hardware and software advances needed for a flight hardware instrument.

Research Task Descriptions: A modular approach for assembling a laser light scattering instrument has been chosen. This allows us to implement dynamic light scattering, static light scattering, dynamic depolarized light scattering, etc. in a manner tailored for each of NASA's Principal Investigators. This allows single angle studies with a simple glovebox apparatus or simultaneous multiple angle studies with a more elaborate rackmount instrument. Dynamic light scattering is used for measuring and correlating fluctuations in the intensity of the light scattered from particles executing Brownian motion. This gives a diffusion coefficient from which particle sizes in a range from 3 nanometers to above 3 microns are derived. Static light scattering -- the measurement of the time-averaged intensity scattered by dispersions of particles and macromolecules (e.g. polymers, proteins, micelles, microemulsions, etc.) -- is an important tool for the determination of particle structure, weight-average molecular weight and particle interactions. Dynamic depolarized laser light scattering examines the weak, horizontally polarized scattered light scattered from a sample and derives dynamical and structural information from this (e.g. - rotational properties and particle aspect ratios).

Progress to Date: User requirements are being assessed through workshops, meetings, and the NASA Research Announcement (NRA) in Fluid Physics. These have indicated/will indicate that critical phenomena, nucleation, spinodal decomposition, gelation, aggregation, diffusion, etc. are influenced by gravity and can be better studied with LLS in a microgravity environment. These basic science questions and many others requiring microgravity and LLS for their answer have been identified by potential Principle Investigators (PIs).

A laptop computer containing a prototype single card correlator capable of both exponential and linear time sampling is being evaluated in our laboratory. Miniature laser and detector modules will arrive and be tested before the end of December 1991. In comparison to the bulk instrumentation that is being replaced, these solid state miniaturized modules are more power efficient, advanced, rugged, reliable, etc. and the problems associated with aligning their traditional counterparts will be obviated through the use of fiber optics.

Fiber optic probes, developed for this project and patented by H.S. Dhadwal from SUNY at Stony Brook, will allow LLS to be used in a concentrated solution (one which does not allow a laser beam to pass through it) for the first time. With back scatter probes, we have been able to study milky concentrations ranging up to 10% weight concentration (for 30 μm particles) without multiple scattering problems. We (Ansari and Dhadwal in particular) have also used these probes to noninvasively track protein aggregation (12 to 120 μm) by observing thermally induced cataracts in bovine eye lenses which have been surgically removed. A multiple fiber optic probe, patented by Dhadwal, has been tested. It allows us to take dynamic light scattering data at low, medium and high angles without needing an index matching fluid around the sample cell to keep the detector from seeing light scattered from the walls of sample cell.

An optical masking technique has been implemented by H.M. Cheung from the University of Akron. It allows data to be taken at any angle (even below 5 degrees with small 5mm diameter sample cells) without index matching fluid. Patents have been issued for this idea (Patent No. 5,028,135 issued July 2, 1991) and a follow-on patent has been applied for. The follow-on idea should allow us to implement dynamic depolarized laser light scattering at zero degrees where rotational and translation diffusion coefficients are decoupled (i.e. - only the rotational diffusion coefficient remains).

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Vibration Isolation Technology (VIT) Advanced Technology Development (ATD)

Universities Space Research Association
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Objectives: The objectives of this project are to: (1) computationally determine vibration sensitivity of Protein Crystal Growth Experiments; (2) determine if these experiments can benefit from vibration isolation techniques; and (3) provide realistic requirements for vibration isolation technology.

Research Task Description: Materials processing in space based laboratories has already yielded higher quality crystals during previous space flights and opportunities for several fluids experiments are anticipated during the extended duration missions planned for the future. Crystal growth in space benefits not only from its reduced gravity environment but also from the absence of the hydrostatic pressure which assists certain crystal growth and refinement methods. Gravity driven phenomena are thus reduced in strength and a purely diffusive fluids behavior can be attained. In addition, past materials science experiments have shown that microgravity can also help produce larger crystals. While gravity related effects are definitely curtailed in space, they are nevertheless present to some degree due to the acceleration environment on board the spacecraft. This residual acceleration level is comprised of quasi-steady, oscillatory and transient components and are caused by a variety of mechanisms. For example, the orbital period of the spacecraft produces low frequency disturbances and the operation of machinery, control thrusters, solar panels, human activity etc., contribute to higher frequency accelerations. These disturbances are collectively referred to as g-jitter and they can be deleterious to certain experiments where the minimization of the acceleration level is important. Advanced vibration isolation techniques can be utilized to actively filter out some of the detrimental frequencies and help in obtaining optimum results. However, the successful application of this technology, requires the detailed analysis of candidate fluids experiments to gauge their response to g-jitter and to determine their acceleration sensitivities.

Several crystal growth experiments in the Protein Crystal Growth (PCG) area, besides others, are expected to be carried out on future shuttle flights and on Space Station Freedom. The need for vibration isolation systems or components for microgravity science experiments can be expected to grow as experiments and available hardware becomes more complex. This technology will also find increased application as the science community develops an awareness of their specific needs relative to the environment available in manned space missions. The development of a tolerable microgravity environment requirement, requires the knowledge of tolerance limits on the allowable g-level and then provide the required technology to achieve this goal. Code SN has recognized the need for this development and initiated an Advanced Technology Development program to address this need. Marshall Space Flight center provides the analysis support for the numerical modeling of different crystal growth methods. This effort will assist in establishing the tolerable acceleration levels for specific fluids experiments. Primary effort is directed towards modeling PCG with the analysis of solution crystal growth scheduled for a later time.

Task 1: PCG Familiarization

Review of protein crystal growth methodology and all past shuttle experimental efforts. Involves the detailed search for PCG information to better understand the current experimental methodology, setup, time-line, difficulties, inflight anomalies, etc. and to plan out a detailed computational approach. This research into past efforts will lead to a better understanding of hardware changes over the past eight or so STS experiments, and a more comprehensive look at the investigators early concern for expected acceleration disturbances. A candidate protein will be chosen for subsequent analysis and modeling and details properties will be compiled for this protein.

Task 2: Code Development and Verification

This effort aims to complete modifications to the 2-D finite difference Navier-Stokes equations solver by introducing the species equation. The modifications will also include the addition of source terms to the momentum equations to account for the solutal buoyancy effects. The code will be thoroughly verified against well documented bench mark solutions. Both steady and time accurate unsteady calculations will be verified. Numerical aspects like code stability, adequacy of grid refinement, numerical diffusion and convergence rate will be investigated.

Task 3: Order of Magnitude Analysis (OMA)

This work will aim at obtaining preliminary estimates of fluid sensitivity to g-jitter using scale analysis. The effect of a single frequency disturbance will be gauged by a simplified analytical model and g-tolerance levels will be estimated. The OMA estimates will provide a first estimate of the sensitivity levels. These estimates will be later checked against results from the detailed numerical computations.

Task 4: Numerical Modeling of PCG

The response of the candidate protein, identified in task 1, will be determined from numerical simulation of different g-jitter scenarios. Simplified boundary conditions will be initially examined followed by more realistic boundary conditions. This task will investigate the fluid response to the following:

- (i) Steady state g-levels, ranging from

$$g = g_0 \text{ (terrestrial gravity)}$$

$$g = 10^{-4}g_0 \setminus$$

=> Microgravity

$$g = 10^{-5}g_0 /$$

$$g = 0 \text{ (zero gravity)}$$

- (ii) Transient excitations: single and multiple step functions. These excitations model random impulse type disturbances that occur on a space based laboratory. Different impulse strengths and durations will be investigated.

- (iii) Periodic accelerations: Simulations will be carried out for low (10^{-3} Hz), intermediate (10^{-2} to 0.1 Hz) and high frequency (> 1 Hz) disturbances. Different amplitudes will be examined. Comparisons will be made with OMA estimates.

- (iv) Combined excitations: In this effort, realistic g-jitter will be simulated by combining transient, residual (low frequency) and oscillatory (intermediate and high frequency) disturbances. Fluid sensitivity will be determined and tolerable g-levels will be established.

Continuous feedback will be provided to NASA Lerc during the entire effort. This will enable identifying critical g-jitter frequencies for a particular experiment and help in the hardware setup of VIT. Tasks 1-4 are scheduled for completion in FY 1991.

Task 5: Numerical Modeling of other Proteins

Among the twenty or so proteins that have flown on previous shuttle flights, several have failed to yield optimum results. Some of these proteins will be examined to see if they will benefit from vibration isolation.

Task 6: Numerical Modeling of other Crystal Growth Processes

Other crystal growth methods will be examined. The growth of crystal from solution (FES) is a good candidate in this regard. The technique will be examined and possibly tasks 1-5 will be undertaken for this crystal growth method. The details pertaining to this task remains to be finalized at a later time.

Progress to Date: Tasks 1-3 and a major portion of task 4 have been completed. Preliminary results from the research effort were presented at the International Workshop on Vibration Isolation Technology for Microgravity Science Applications, April 23-25, 1991, in Cleveland, Ohio. Feedback from the international forum was incorporated into the ongoing investigation. An overall research status and progress was also outlined during the ATD Program Review in July 1991 at NASA headquarters.

The salient results from the investigation are as follows:

1. G-jitter dominates the spacecraft acceleration environment.
2. It is comprised of a myriad frequencies and displays no preferred orientation.
3. The g-jitter magnitudes can be as high as 1 micro-g (10^{-3} g).
4. Impulse type disturbances are random in nature; It is prudent to take remedial measures.
5. PCG observations and analyses indicate susceptibility to g-jitter.
6. Calculations show the PCG flow field to be susceptible to 1-10 Hz frequency range.
7. Impulse type disturbances are also deleterious to PCG (drop dislodgment, multiple crystals etc. are caused by such impacts).
8. PCG is a PRIME candidate for Vibration Isolation. NASA Lerc developed active vibration isolation system filters up to 0.1 Hz; Passive systems can also filter down to this frequency level.
9. Passive Isolation System for PCG - An obvious FIRST RECOMMENDATION.

The most recent results from the study were presented at the 'Fourth International Conference on Crystal Growth of Biological Macromolecules', August 18-23, 1991 in Freiburg, Germany. A comprehensive paper summarizing the results is under preparation for the Journal of Crystal Growth.

Publications

Ramachandran, N., "G-Jitter Convection in Enclosures," *9th Intl. Heat Transfer Conference*, August 19-24, 1990, Jerusalem, Israel.

Ramachandran, N., "Thermal Buoyancy and Solutal Convection due to G-jitter," *AIAA 29th Aerospace Sciences Meeting*, Reno, 1991.

Ramachandran, N., "Convection Effects and Vibration Isolation for Protein Crystal Growth," *Fourth Intl. Conference on Crystallization of Biological Macromolecules*, August 18-24, 1991. Manuscript under preparation for Journal of Crystal Growth.

Advanced Technology Development Transparent Furnace Technology

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Objectives: Our project objectives are; a) development of a multizone transparent furnace system, b) development of multivariable, adaptive control schemes for multizone transparent furnaces, and c) development of an image processing system that will be used to quantify the interfaces during crystal growth experiments.

Research Project Description: Transparent furnaces allow for real time observation of the material sample and provide flexible viewing of the material processing phenomena. This viewing may be enhanced by lenses, cameras, and other devices. The applications of transparent furnaces include the study of low temperature phase change model materials such as succinonitrile, water, ammonium chloride/water, and higher temperature practical materials such as Lead Chloride, Cesium Iodide/Thallium Iodide, and Cadmium Telluride.

Previous research in the crystal growth community has established that the shape of the solid-liquid interface is a function of the imposed temperature profile, translational speed of the ampoule, and the physical properties of the crystal growth material and ampoule. It is believed that a horizontal and planar interface reduce crystalline imperfection due to the reduction of thermal stresses when the interface is flat.

Since the shape of the interface is a significant characteristic of this process, automatic detection and quantification of this variable can play an important role in crystal growth. The effects of temperature gradient and ampoule translation speed may be observed by comparing images obtained from different experiments. The crystal growth rate and the periodic movement of the interface due to ampoule end effects and various other disturbances also can be observed and recorded. Interface shape information obtained from such experiments may eventually be used as feedback signal for interface shape control.

Rapid progress in understanding the effects of convection on crystal growth in enclosed ampoules is only possible through simultaneous and coordinated theoretical, numerical and experimental investigations. To solve equations describing the coupling among heat, momentum, and mass transfer in enclosed ampoules, boundary conditions are needed. Naturally, if successful comparison between numerical and experimental results is intended, these boundary conditions have to be determined accurately. Under these circumstances, it appears that direct measurement of boundary conditions during experimental studies would provide useful information to numerical modeling efforts and eventually help the experimentalist. Transparent furnaces provide the capability to obtain such boundary conditions via experimental measurements.

Due to inevitable heat exchange between different heating zones and the transient nature of the process, the dynamics of multizone furnaces is time varying, distributed and therefore complex in nature. Single input-single output (SISO) models do not explicitly account for the temperature and powers of neighboring heating zones in a multizone furnace. Changes in power and temperature of one zone act as input to other zones. This phenomena is observed through a non-proportional power distribution along the furnace. The power distribution effects the amount of energy transmitted into the material sample. A multivariable model provides the capability to account for the interaction between zones. By including these interactions explicitly, using multivariable techniques, a multizone furnace controller can be designed to minimize temperature fluctuations within the furnace.

Progress to Date: To date we have developed an eight zone transparent furnace used to study directional solidification and physical vapor transport crystal growth of electronic materials. The furnace is made up of eight individual zones arranged serially in a vertical orientation. Numbering the zone 1 through 8, top to bottom, the zone sizes are as follows: zones 1, 2, 7, and 8 are 75 mm long; zones 3 and 6 are 38 mm long; and zones 4 and 5 are 25 mm long. The inside diameter of all zones is 32 mm. Each zone is made from 1.5 mm thick quartz tube inside of which is wound 20-gauge Kanthal heating wire. The ampoule containing crystal growth material is suspended in the furnace via a Kanthal wire connected externally to a linear stepping motor that has a resolution of 1 micron. Temperature control based on a 16-bit computer system uses a 12-bit A-to-D conversion card for temperatures ranging from 0 to 1200°C. The zone temperatures are controlled by computer to achieve a specific axial temperature profile.

Under this research an eight zone transparent crystal growth furnace has been controlled by two different SISO adaptive controllers. Changes in the furnace dynamics and the interaction between the neighboring heating zones are estimated by an on line Least Squares algorithm. The pole placement and the PID control algorithms have been tested on the growth of Lead Bromide crystals. Both controllers performed according to their expected levels of performance. However it was found that the pole placement controller is much more flexible to implement any desired closed loop feedback dynamics. The adaptive PID controller only executes the Ziegler-Nichols based performance index.

For an explicit model of the heat interactions between heating zones, a MIMO (multivariable) furnace model has also been determined. The multivariable model more accurately represents the dynamics expected in multizone furnaces.

Several different image processing routines have been applied to find the solid-liquid interface while a crystal is growing within a transparent furnace. A technique based on the Heuckel edge model was found the most effective means to determine the position and shape of the interface. This shape information has been used to assess the morphology of a crystal while it is growing.

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C. Batur, R. B. Sharpless, W. M. B. Duval, B. N. Rosenthal, N. B. Singh, "Computer Imaging-Based Detection and Quantification of Solid-Liquid Interface during Crystal Growth," *American Society of Mechanical Engineers, Intelligent Processing of Materials Annual Winter Meeting*, MD-Vol. 21, pp. 39-51, November 25-30, 1990.

R. B. Sharpless, "Intelligent Control of Crystal Growth", Masters Thesis, University of Akron, Akron, Ohio, May, 1991.

W. M. B Duval, B. N. Rosenthal, C. Batur, R. B. Sharpless, N. B. Singh, "Parametric Effects of External Variables on Interface Morphology of Lead Bromide Crystals," Gordon Research Conference on Crystal Growth, Plymouth, NH, July 15, 1991.

Non-Contact Temperature Measurement

Jet Propulsion Laboratory
Dr. Andrew S. W. Thomas
October 1990 - October 1991

Objectives: The long-term objective of this Advanced Technology Activity (ATD) is to develop non-invasive temperature measurement systems for containerless processing of materials in microgravity applications. Specifically this will consist of:

Determining the accuracy with which target radiance can be determined and quantifying the effect of extraneous factors such as window reflectance, wall radiance etc.

Developing methods of performing an in-situ measurement of the target surface properties so that the emissivity may be determined, from which the measured target radiance may be converted to true temperature.

Evaluating the technologies that are developed using heated targets levitated in low-gravity conditions.

Research Task Description: An experimental analysis and theoretical examination of the measurement of true target radiance in a heated-wall furnace, including the effects of window reflections etc. Development of an active laser interrogative system based on reflection changes in polarized laser light from which emissivity can be derived. Development of a two-color laser pyrometer concept that performs measurement of target reflectance at two closely spaced wavelengths from which an emissivity ratio may be determined. Testing of the active pyrometer systems on-board the KC-135 aircraft with samples heated and positioned with electromagnetic and electrostatic levitation systems.

Progress to Date: A mockup of the Dual Zone Chamber of the Drop Physics Module was fabricated with electrical heating to provide a wall temperature of 800°C. A spherical platinum shell, also heated electrically, was used as the target. Radiance data were acquired with and without wall heating, with and without windows, and for different wavelength pyrometer systems. A theoretical examination of the reflection problem was undertaken to reconcile inconsistencies between the experimental observations and conventional paraxial optics theory. These results show the importance of non-paraxial optics to the problem and have been submitted to the Review of Scientific Instruments for publication.

A polarimetric emissivity measurement device was delivered to JPL for evaluation as part of a subcontracted effort. Preliminary tests on a platinum target show sensitivity to alignment and care is needed in setting up the targets if repeatable data are to be obtained. This version of the device does not meet its rated performance as the measurements appear to have only about a 10% repeatability. An advanced version of the device is scheduled for delivery in October 1991 that should provide superior performance.

An effort was subcontracted to initiate the development of a two-color laser pyrometer concept. The prototype device is scheduled for delivery in mid 1992 for evaluation at JPL. As designed, it will perform near-simultaneous measurements of target radiance and target reflectance at wavelengths of 780 and 904 nanometers. From these data a true target temperature may be computed using the measured reflectance ratio to provide the unknown emissivity ratio.

Low-gravity testing of the candidate pyrometer technologies is not scheduled until 1992. However, in the interim the laboratory capabilities established under this activity, and the pyrometer systems in use, have been made available to the other JPL microgravity activities. These include

pyrometer calibrations for undercooling studies in collaboration with Caltech, and transient heat transfer measurements to support laser heating studies.

Publications

Thomas, A. S. W., "Correction-free Pyrometry in Radiant-wall Furnaces for Microgravity Applications," Rev. Sci. Instrum., 1991 (submitted).

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